Hokudai Cosmochemistry

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Development of Isotope-Microscopy and Implications for Cosmochemistry

Hisayoshi Yurimoto, Kazuhide Nagashima
Hokkaido University, University of Hawaii

Why do we need Isotope Microscopy?

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- Isotopes are used as tracers for
  - chemical reaction path, circulation of elements, origin of elements,and dating, etc.
- When solid phases are newly produced in an environment, the environmental change brings with variations of isotope ratios. The time variation are sequentially fixed in the growing solid.
- If we visualize 3-D distribution of isotope ratios in the solid, we learn the evolution of the environment. If the environment is open system, we learn origin of the element.

These are the reason why we need isotope-microscopy.

- However, the isotopic variation in nature is usually small (less than 1%). High precision measurements for isotope ratios are required.
- How to realize such isotope-microscopy?

Isotope Microscopy using SIMS

Natural Cosmochemistry
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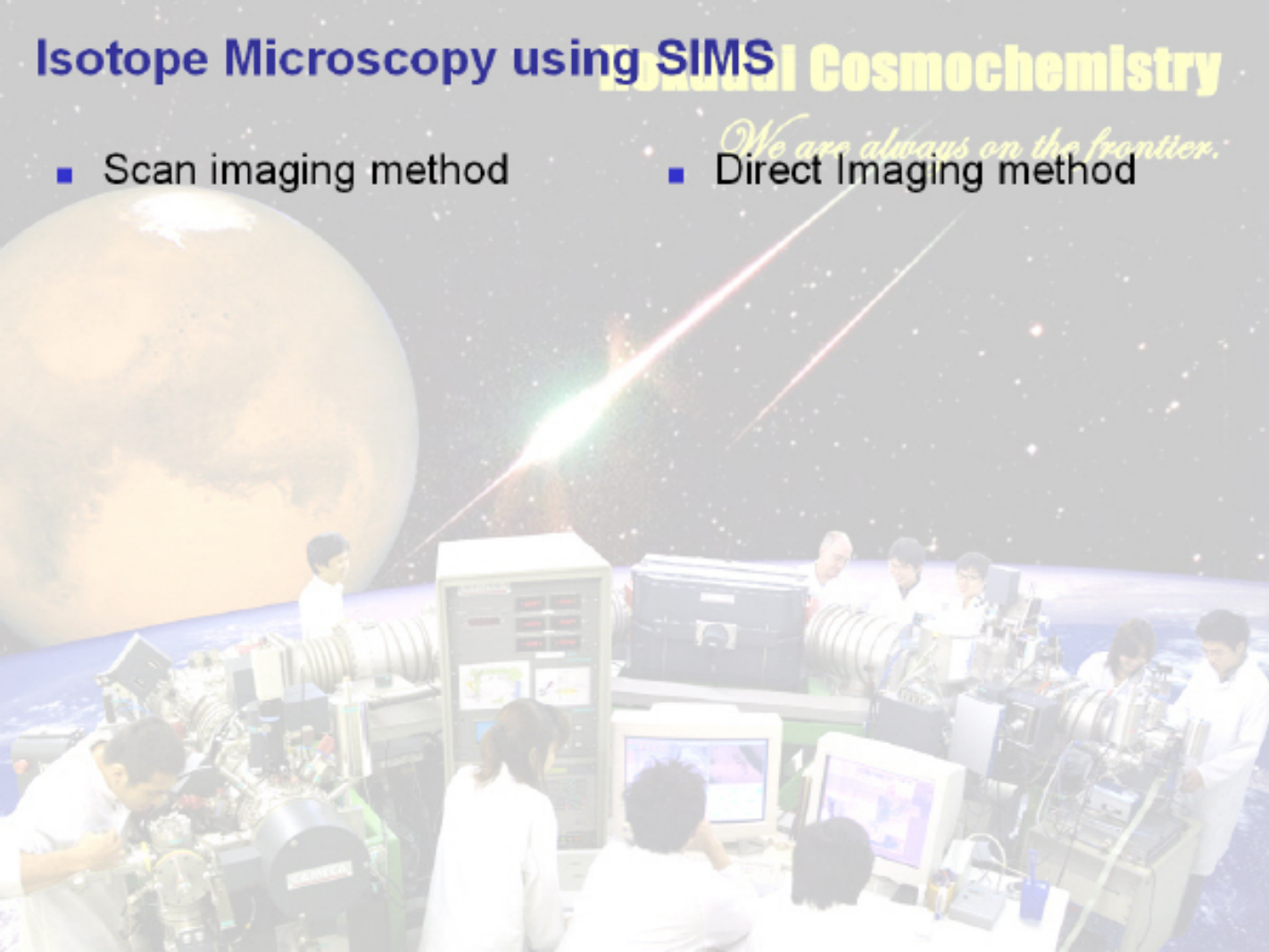
- Scan imaging method
- Direct Imaging method



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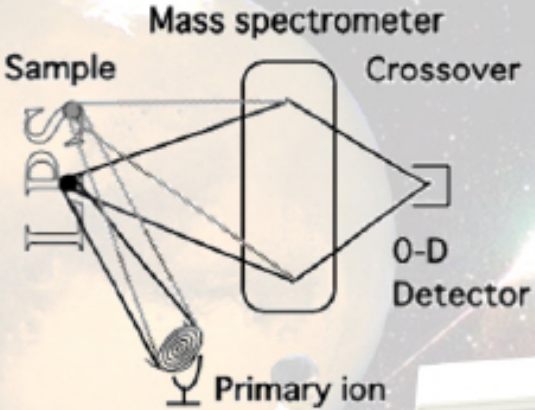

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Terrestrial Cosmochemistry
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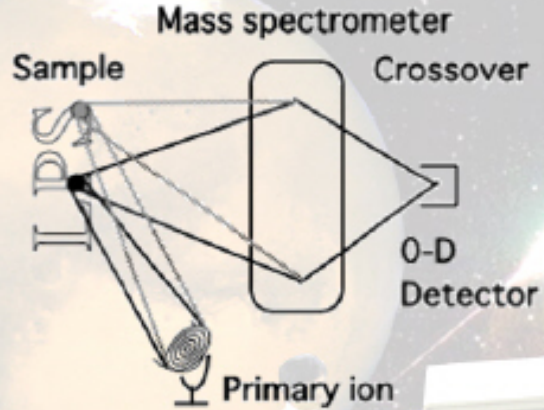

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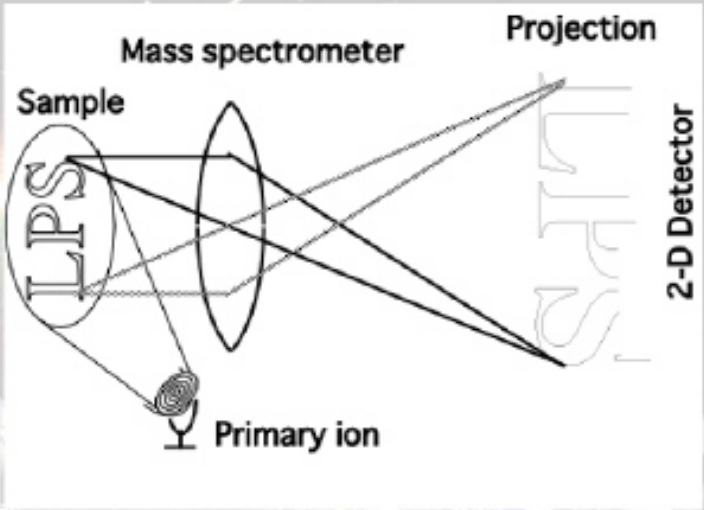

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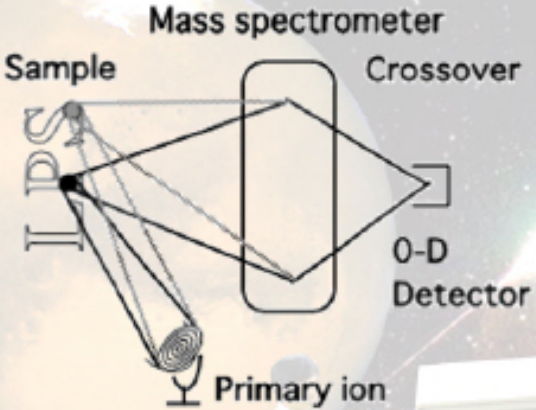

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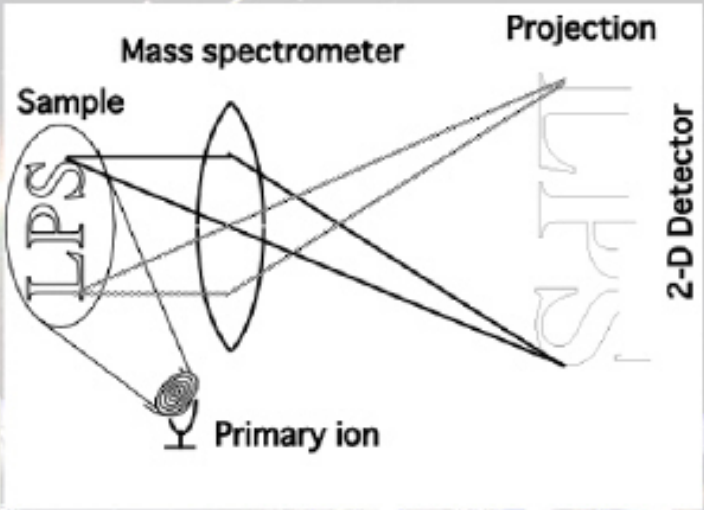

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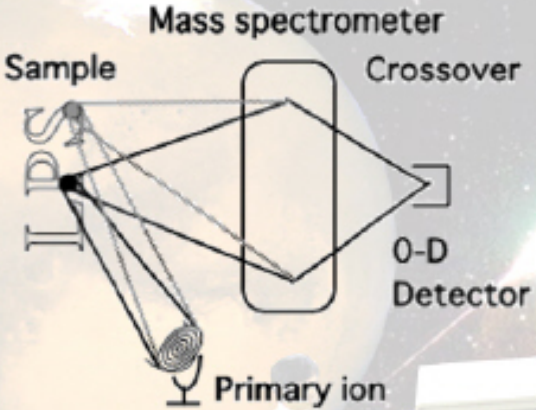

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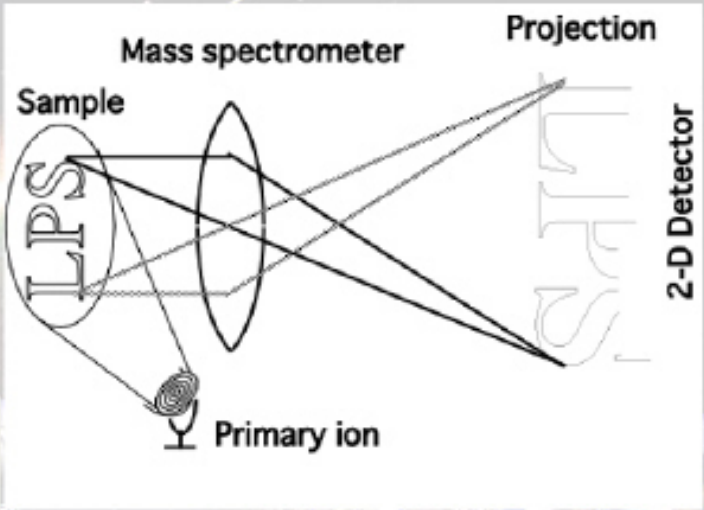

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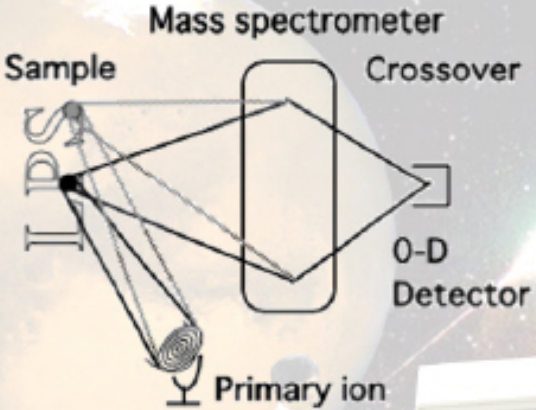

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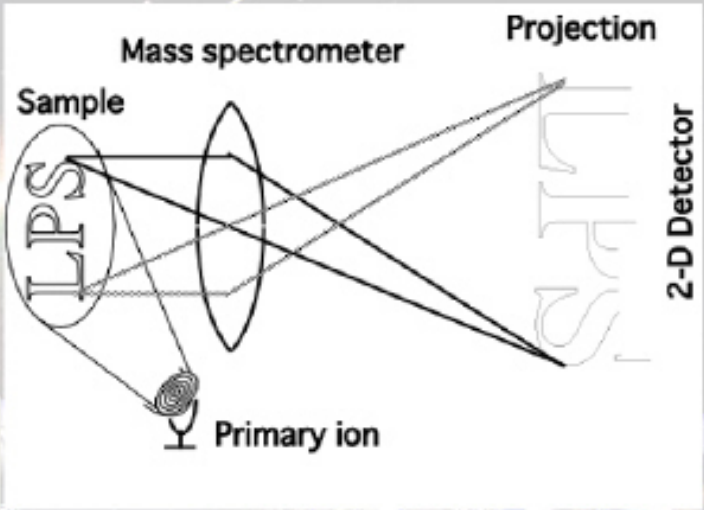

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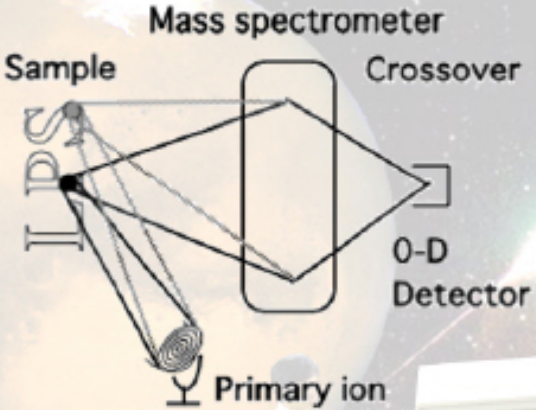

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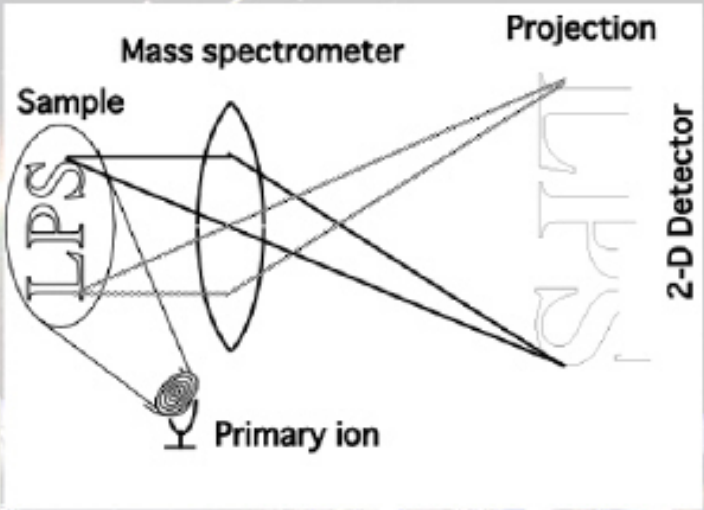

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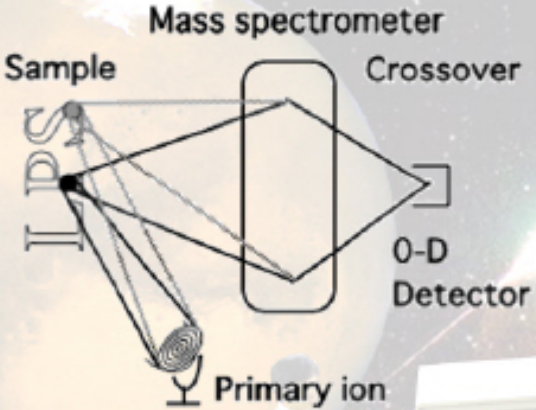

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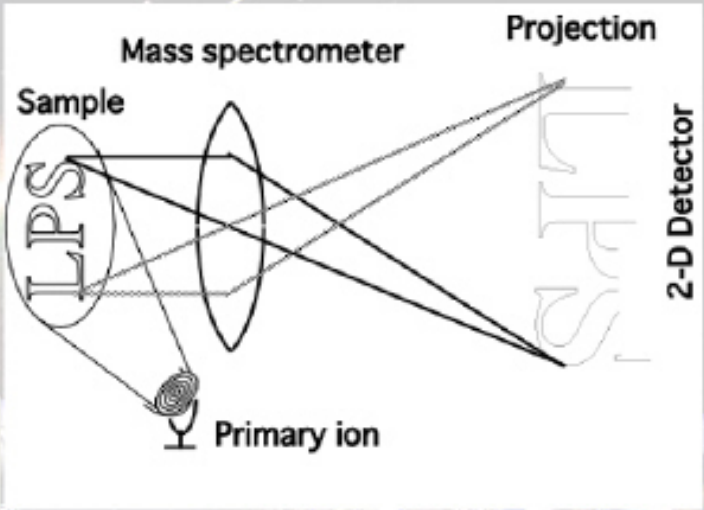

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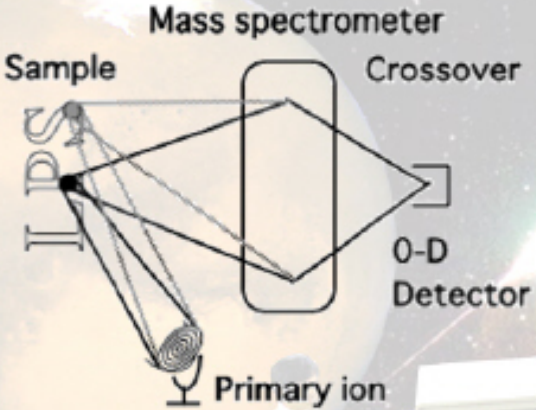

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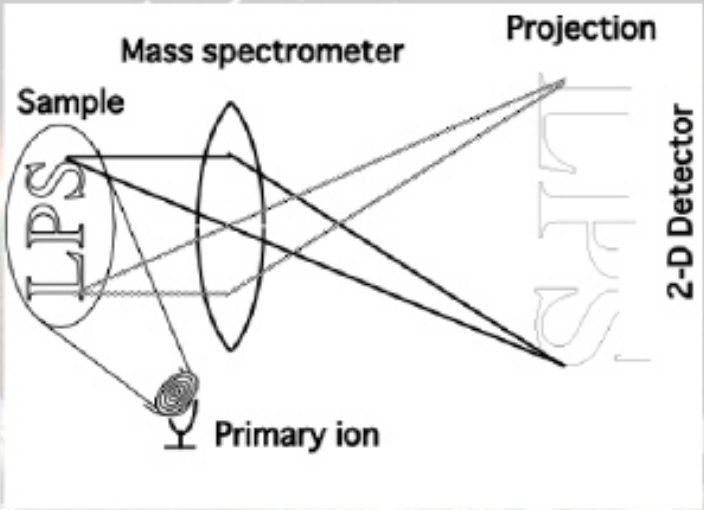

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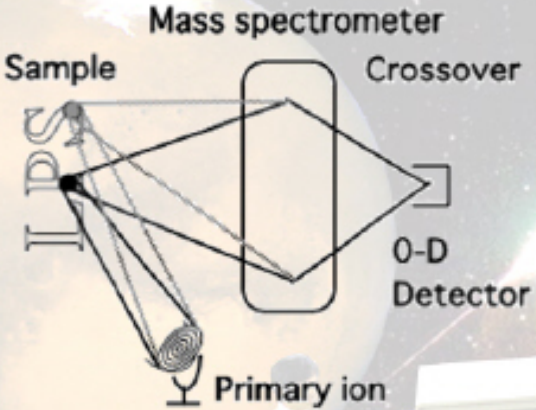

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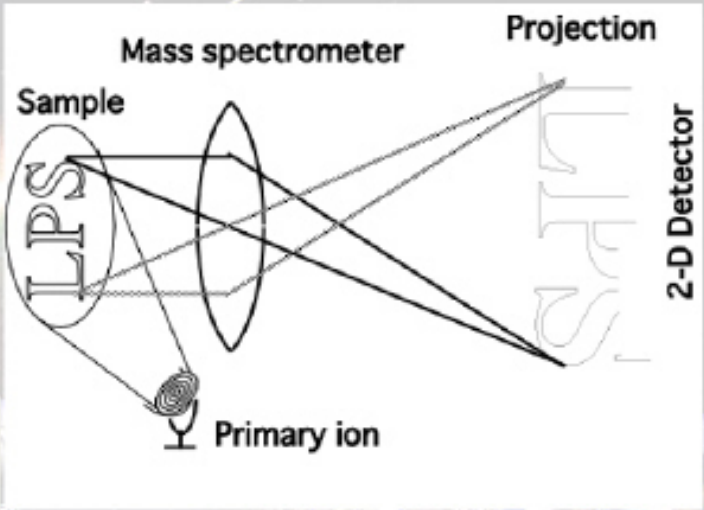

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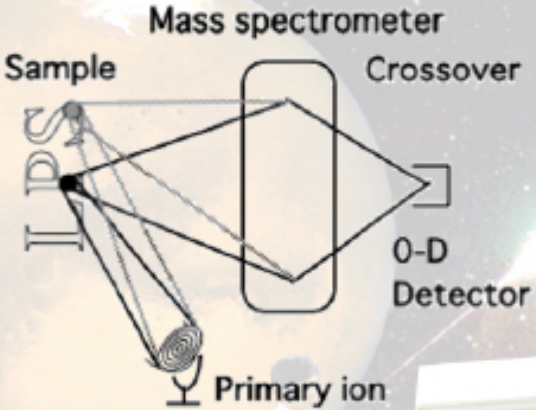

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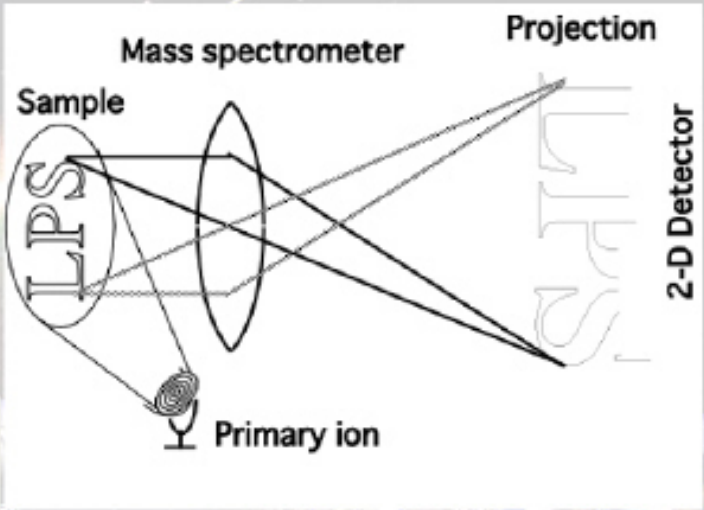

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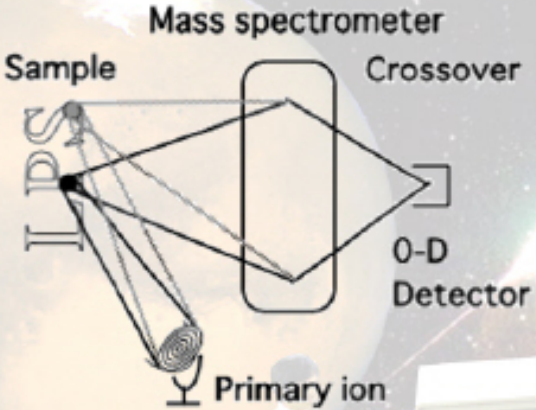

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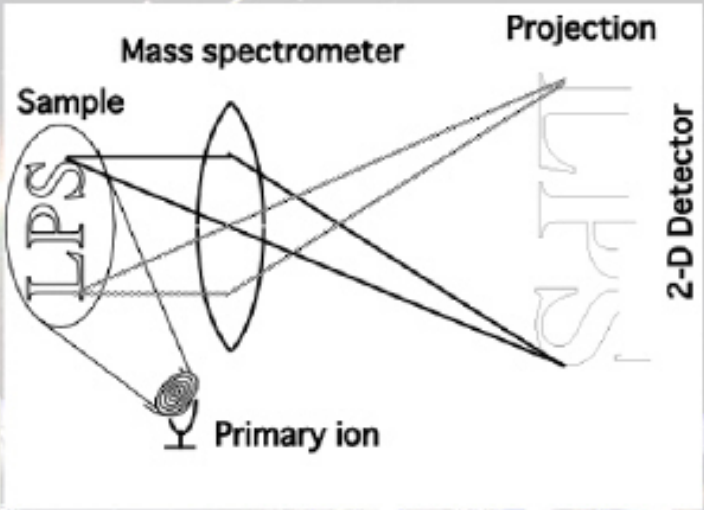

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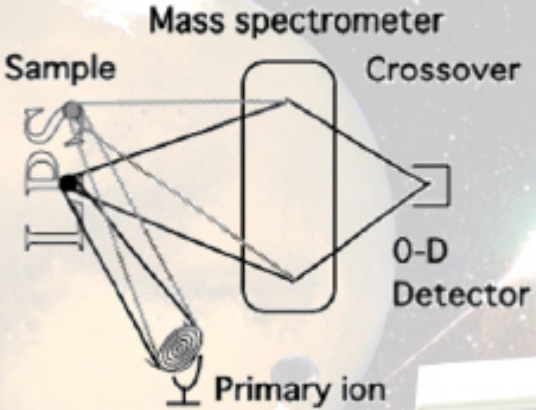

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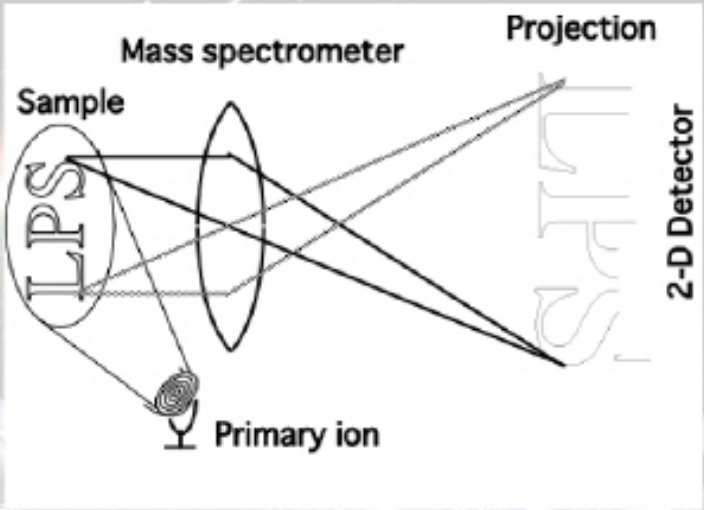

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Isotope Microscopy using SIMS

- Scan imaging method
  - High Spatial Resolution with a fine probe size
  - Long measurement time for wide area
    - High precision analysis is difficult for wide area
- Direct Imaging method
  - Wide Area with High Intensity Signals
    - High precision analysis
  - Spatial Resolution is limited by ion optics

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Ion Optics of Direct-Imaging (Stigmatic) SIMS

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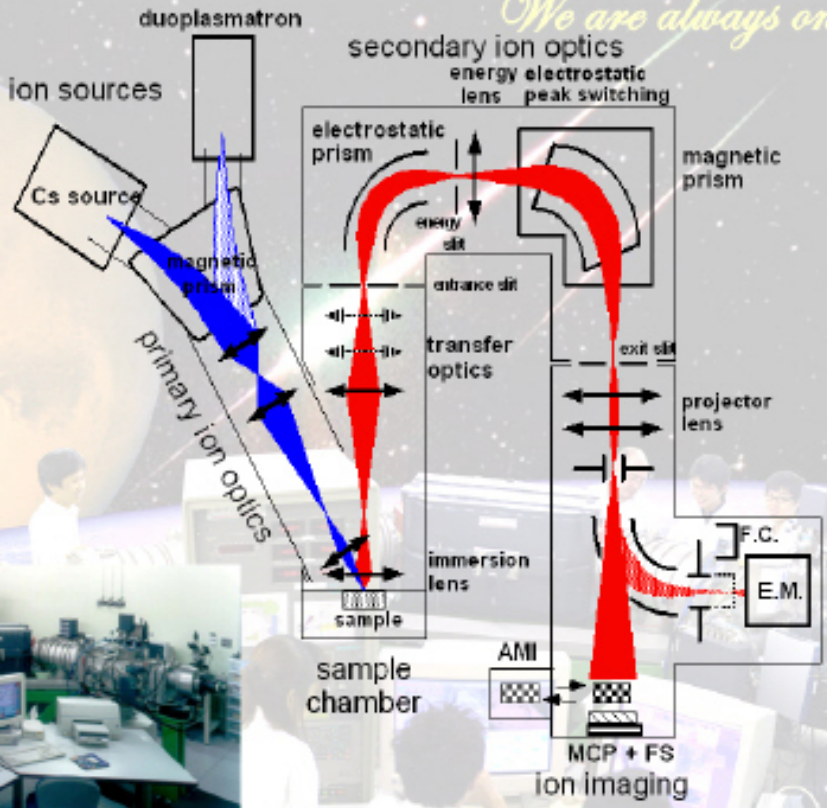

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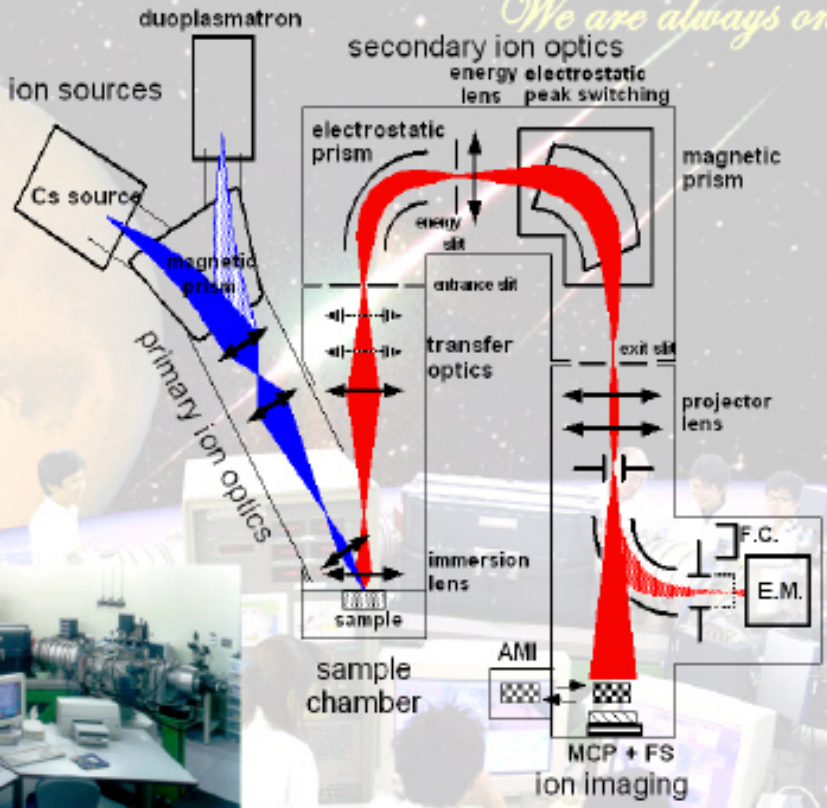

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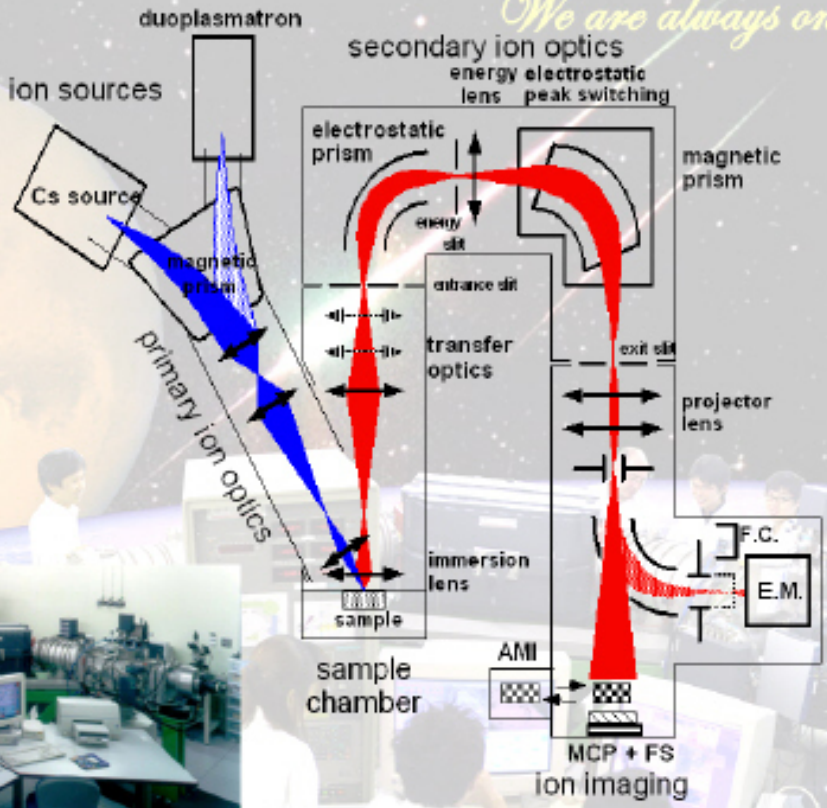

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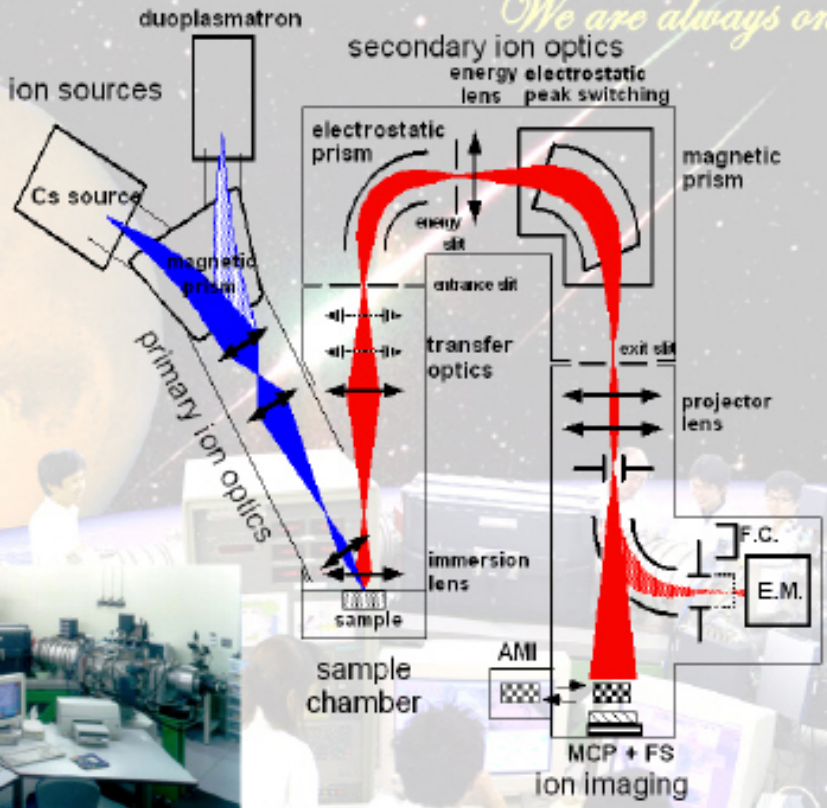

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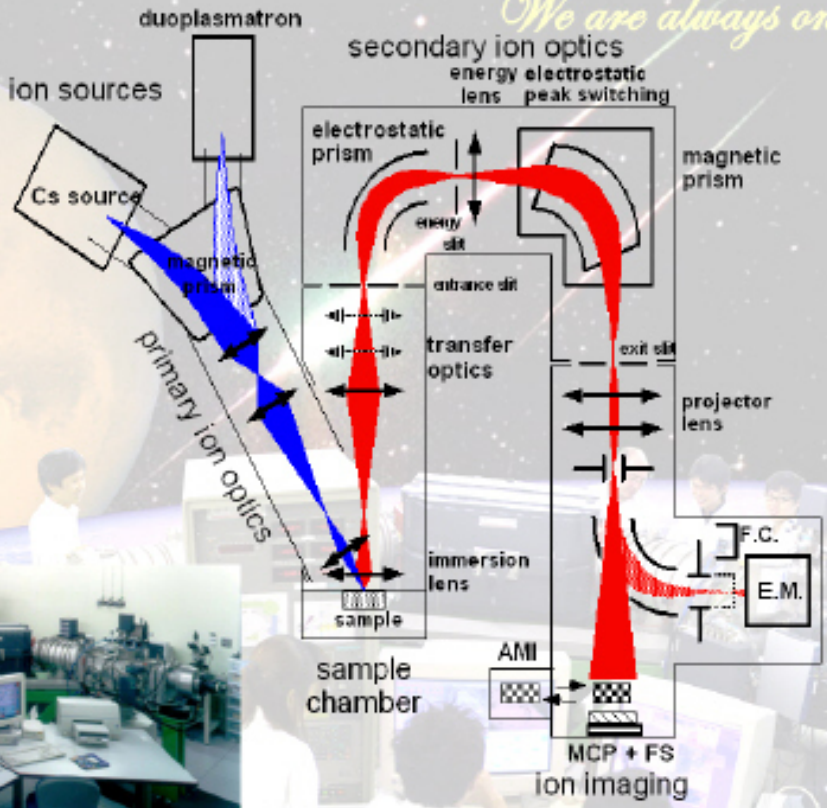

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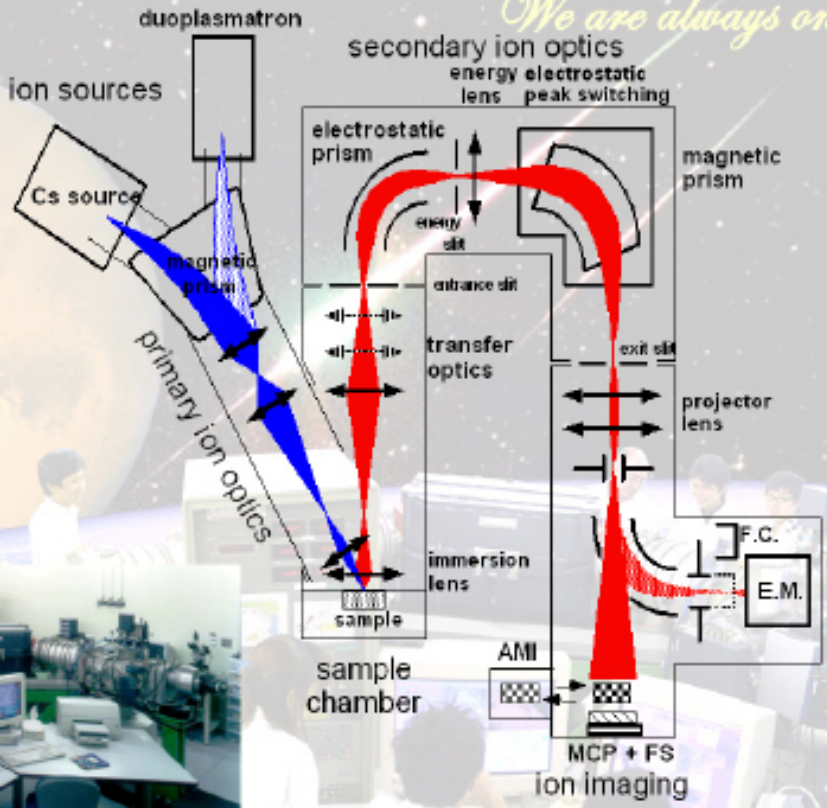

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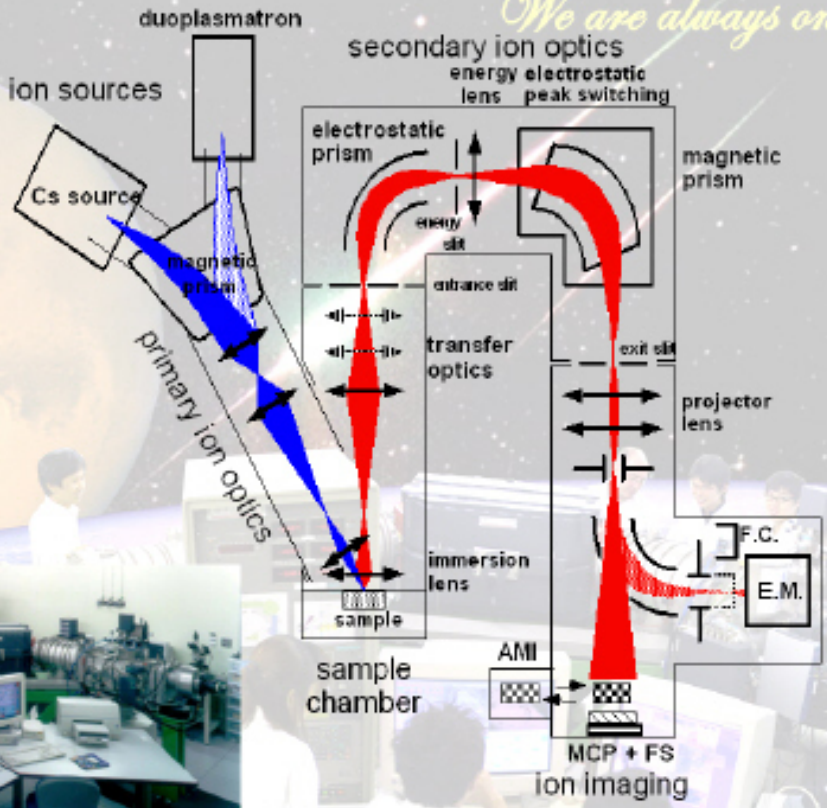

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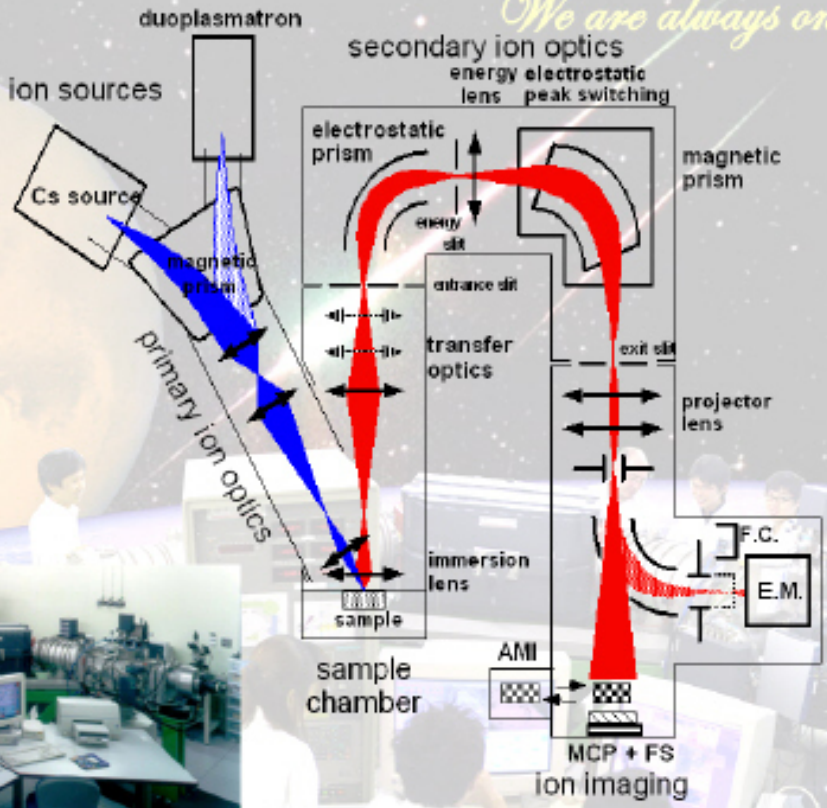

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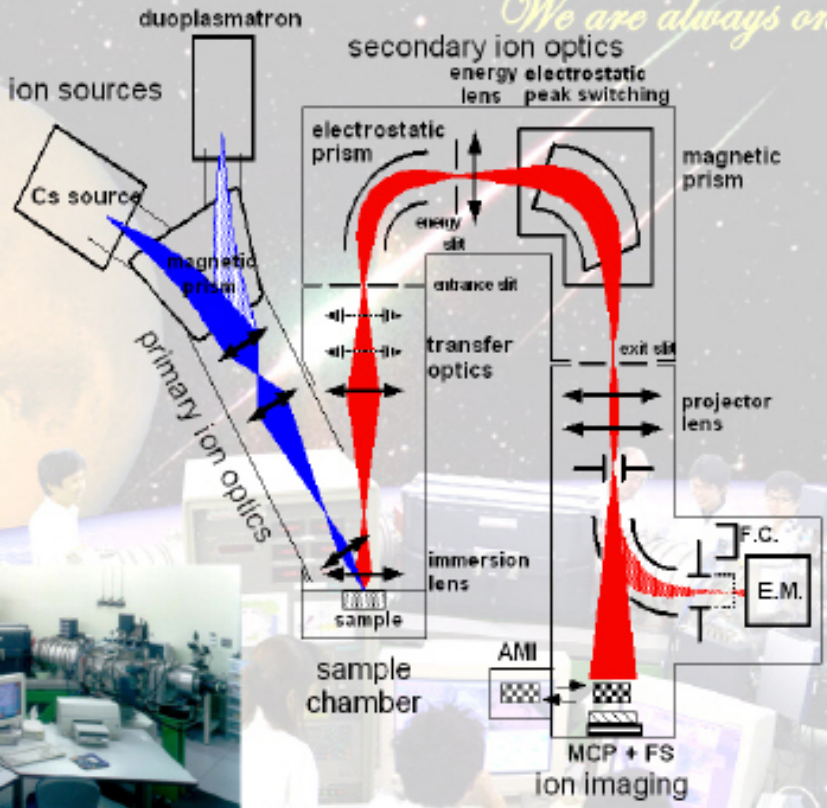

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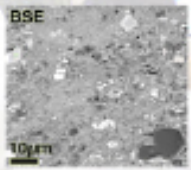

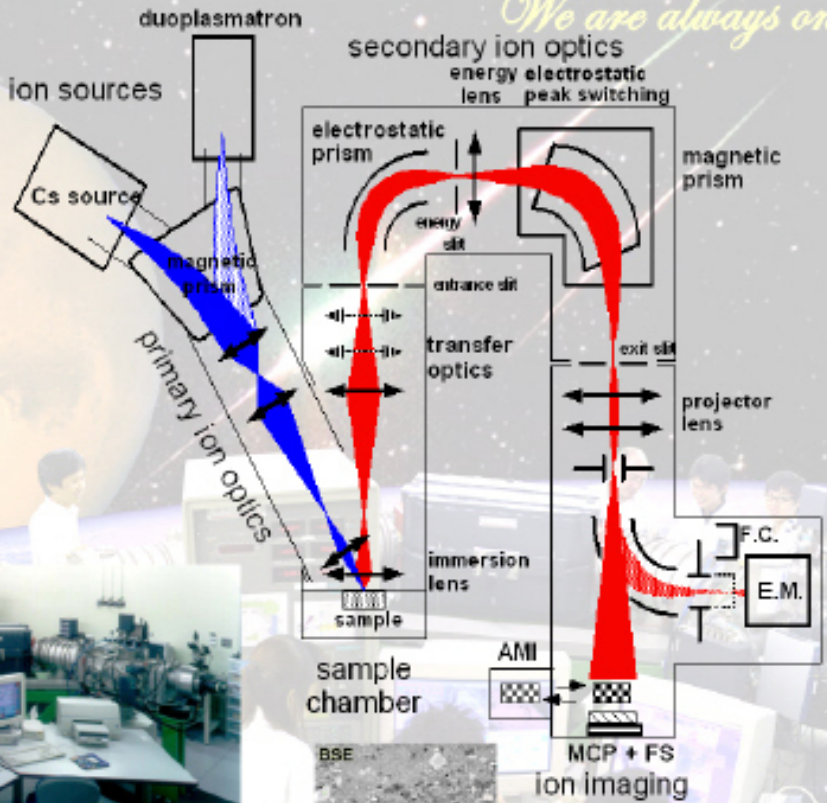

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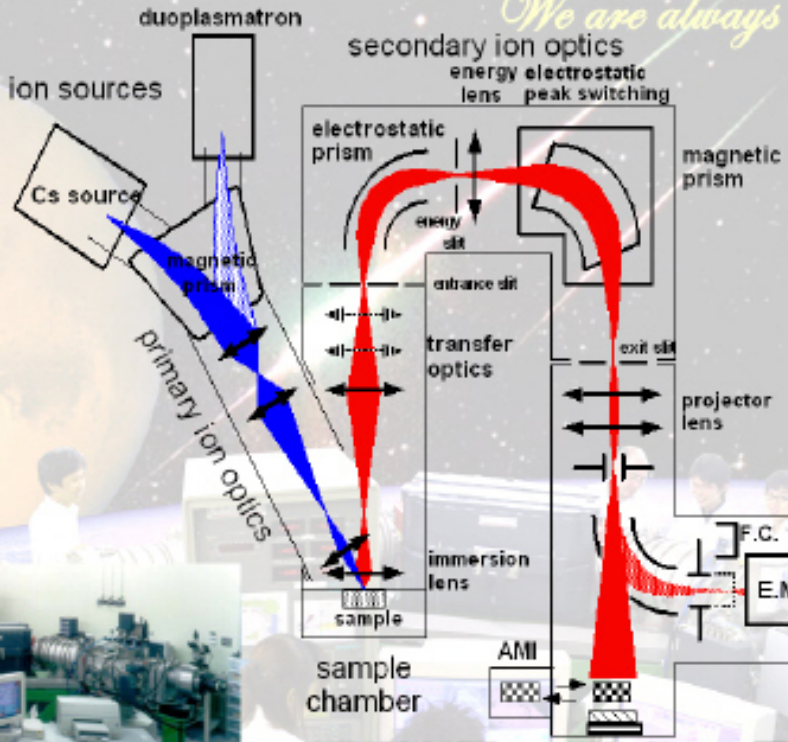

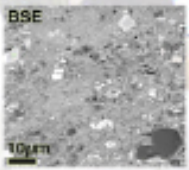

Spatial Resolution of Stigmatic Optics

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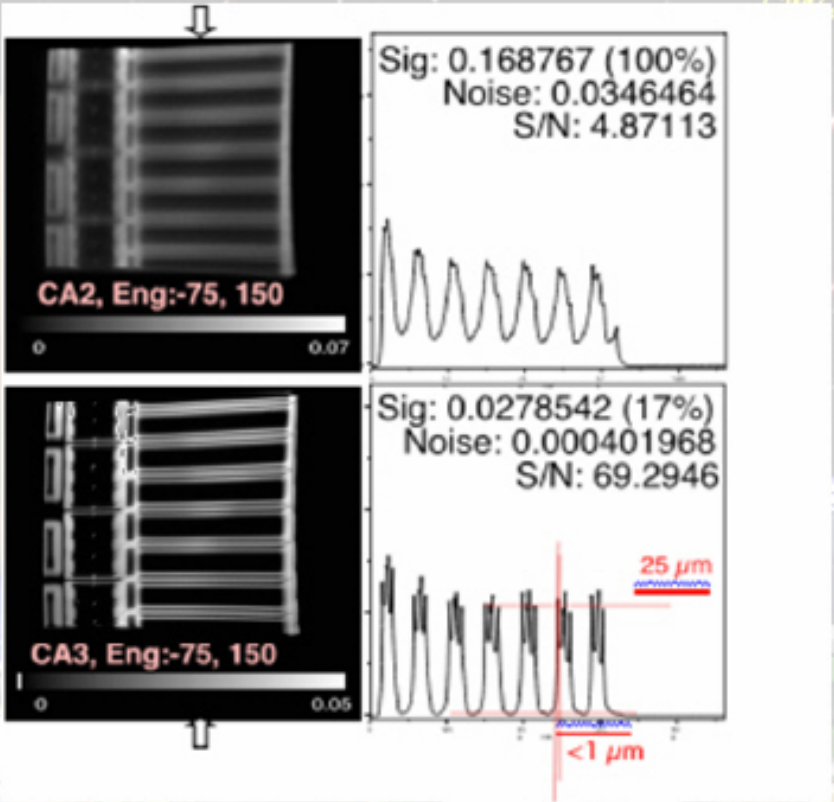

^{16}O / cps
(pri: 0.1nA)

CA #2 (150µm)
2.3×10^7

CA #3 (50µm)
3.3×10^6

Cameca ims1270

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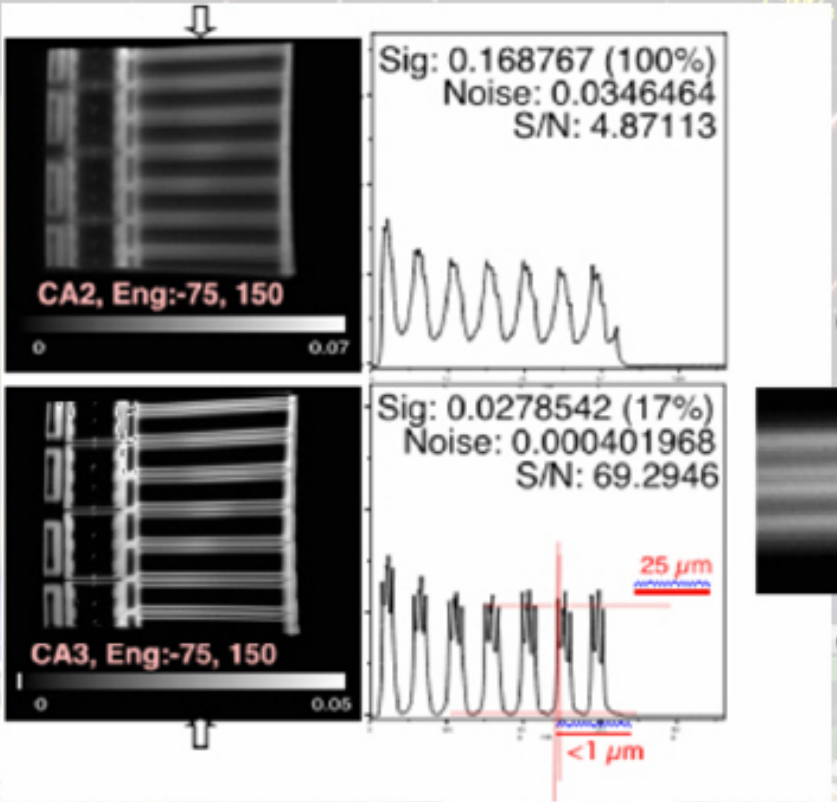
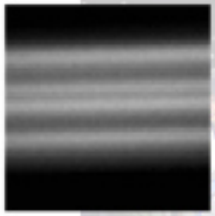


^{16}O / cps
(pri: 0.1nA)

CA #2 (150µm)
2.3×10^7



CA #3 (50µm)
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Cameca ims1270

Spatial Resolution of Stigmatic Optics

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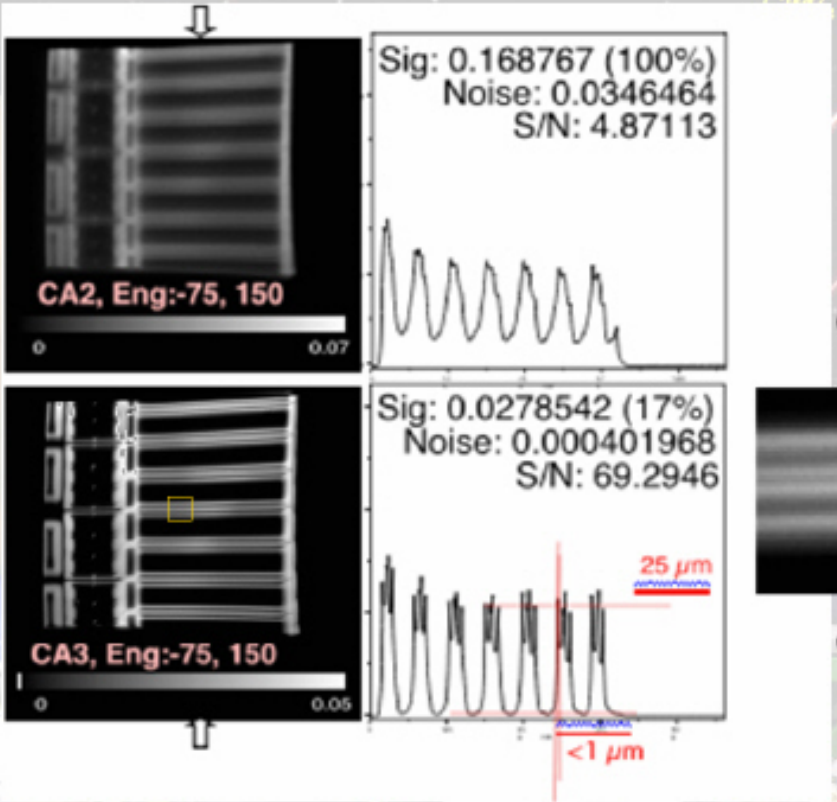

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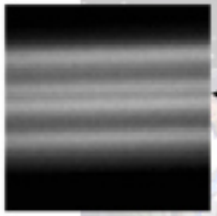

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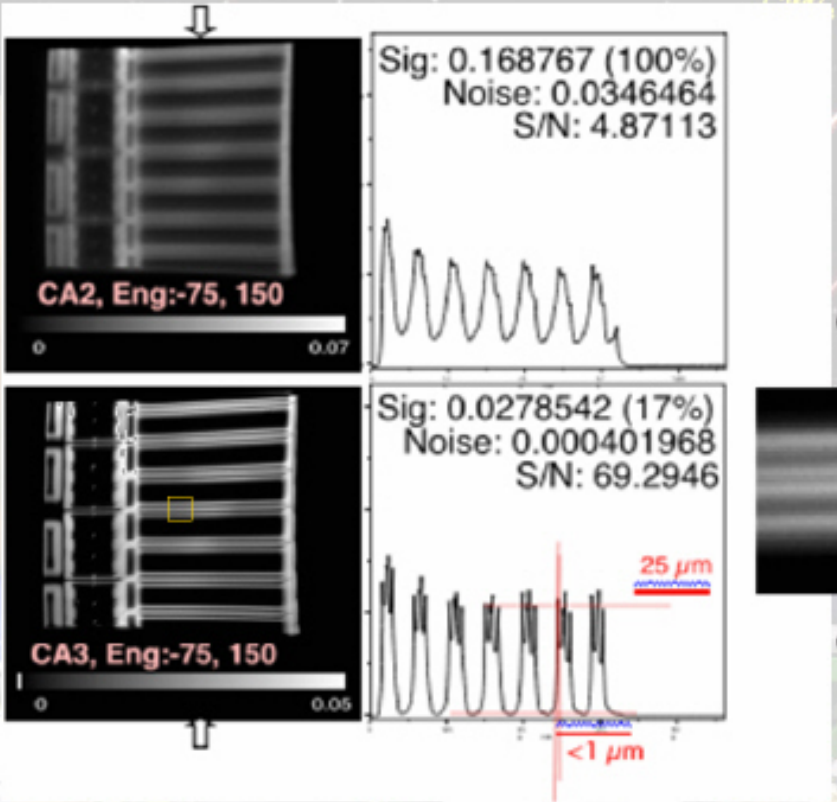

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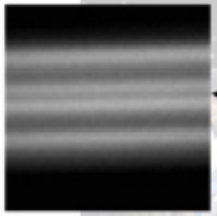

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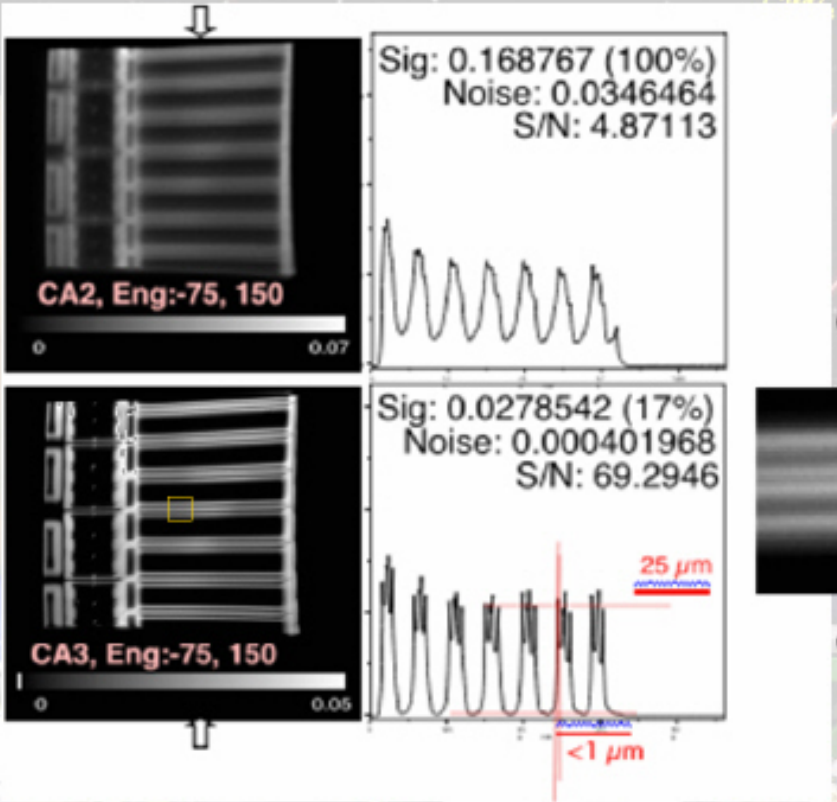

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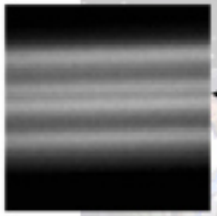

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Cameca ims1270

Performance of Secondary Ion Optics for Cameca ims 1270

Nokadar Cosmochemistry
We are always on the frontier.

- Spatial resolution: sub-micro meter
- Statistical precision: ~0.1% (1‰) for ~10 nm sputtering depth
- Image field: 200 x 200 μm for conductors
  100 x 100 μm for insulators
- Requirements for ion-imaging detectors
  - $>10^6$ counts/(0.5 μm)^2/(10 nm)
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Conventional 2D-Detectors for Stigmatic SIMS

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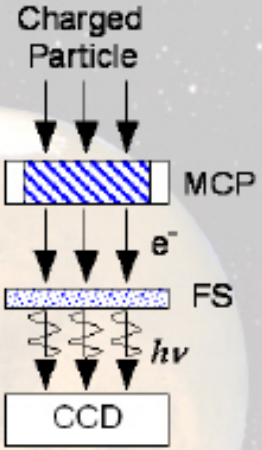
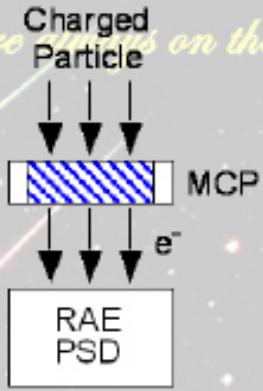


- MCP+FS+CCD system
  - Poor Linearity
    - FS
  - Narrow Dynamic Range
    - FS
  - Robustness: poor
    - FS, MCP



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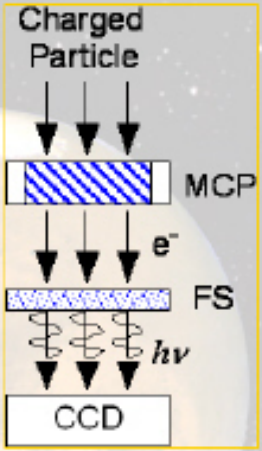

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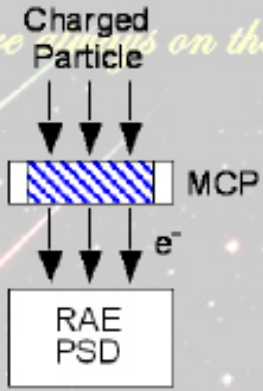

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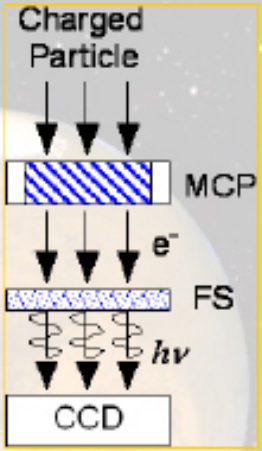

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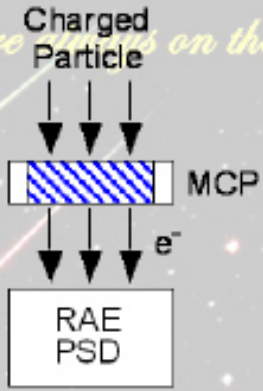

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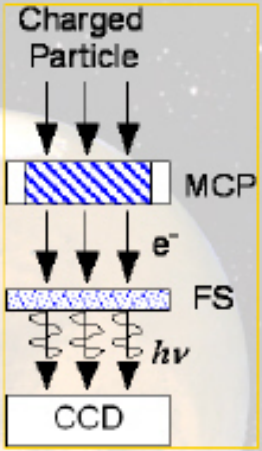

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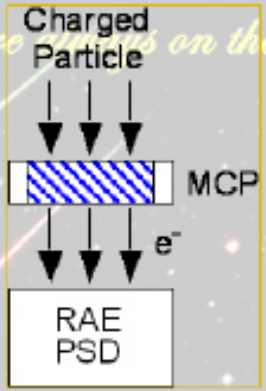

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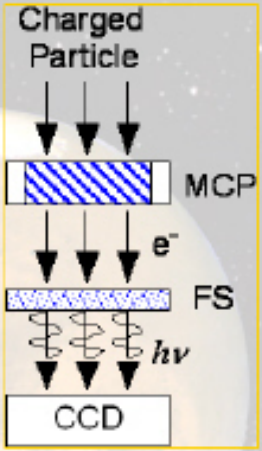

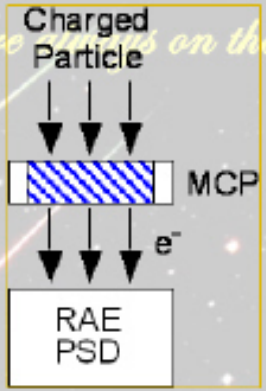

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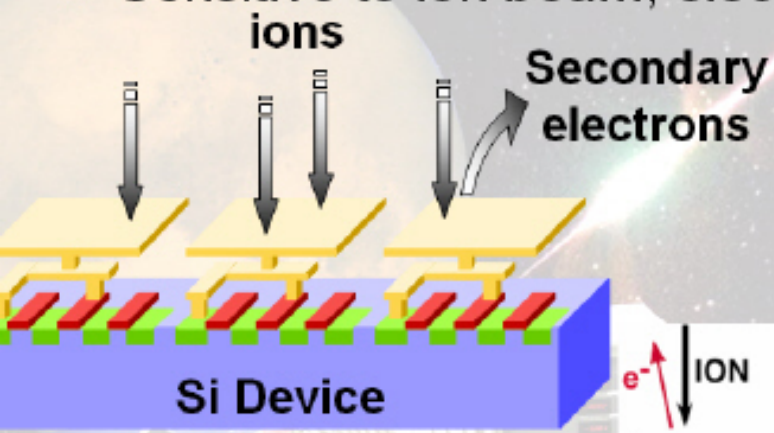
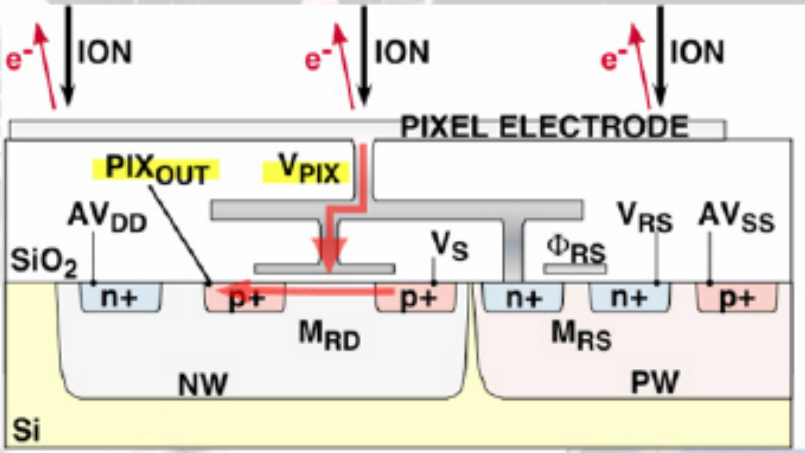
SCAPS (Stacked CMOS Active Pixel Sensor)

Nuclear Semiconductor
We are always on the frontier.

- CMOS imager
- Sensitive to ion beam, electron beam and photon





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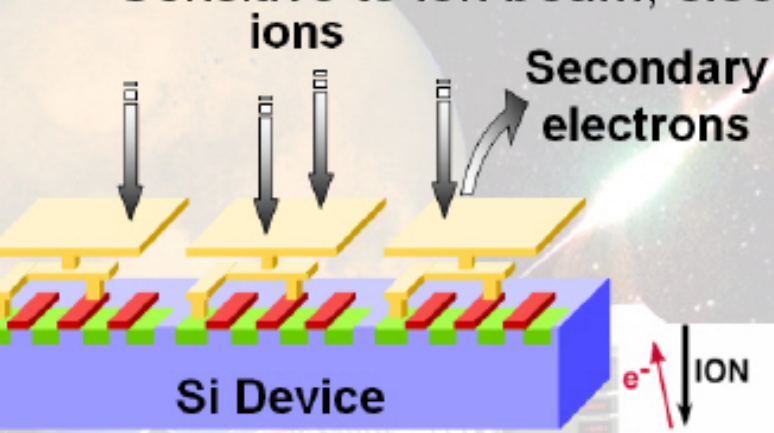

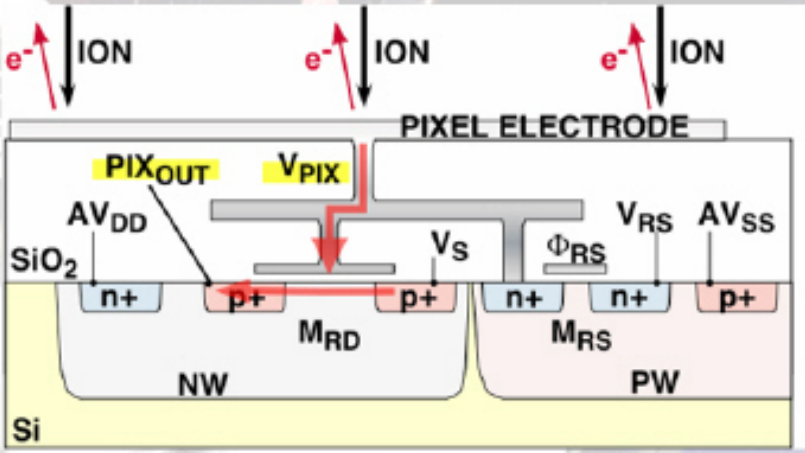

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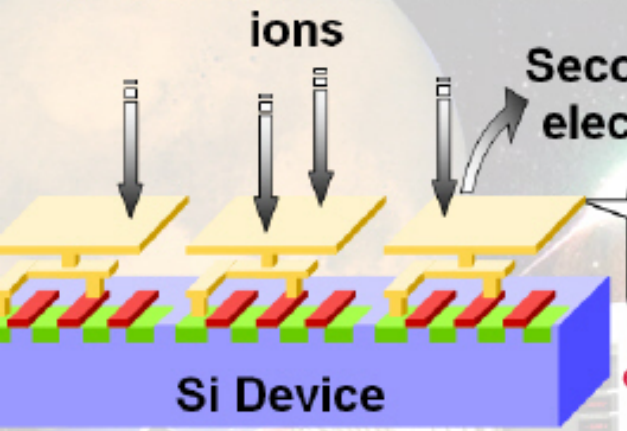

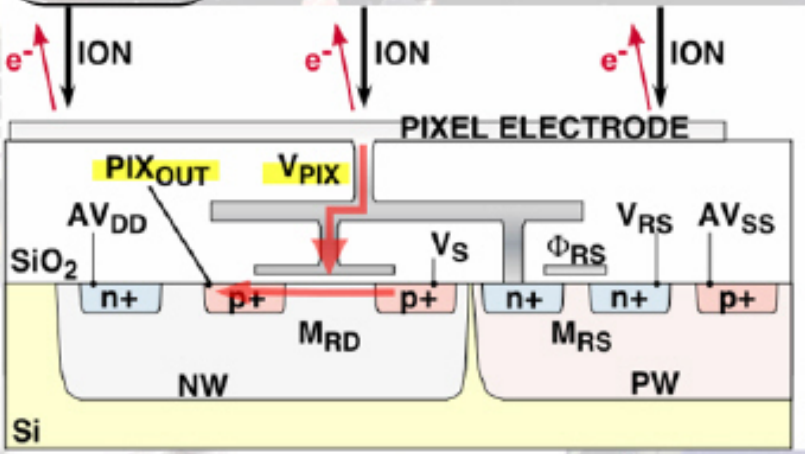

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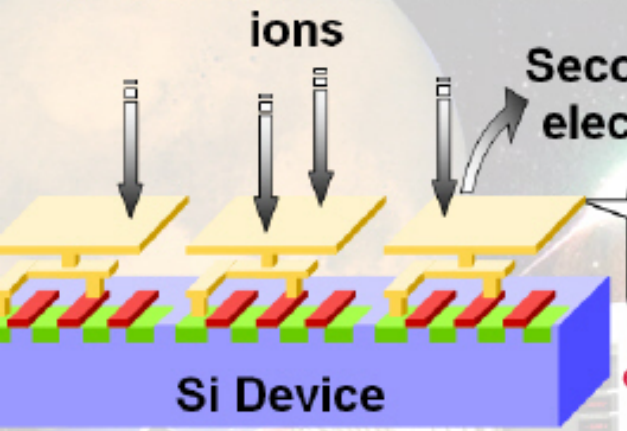

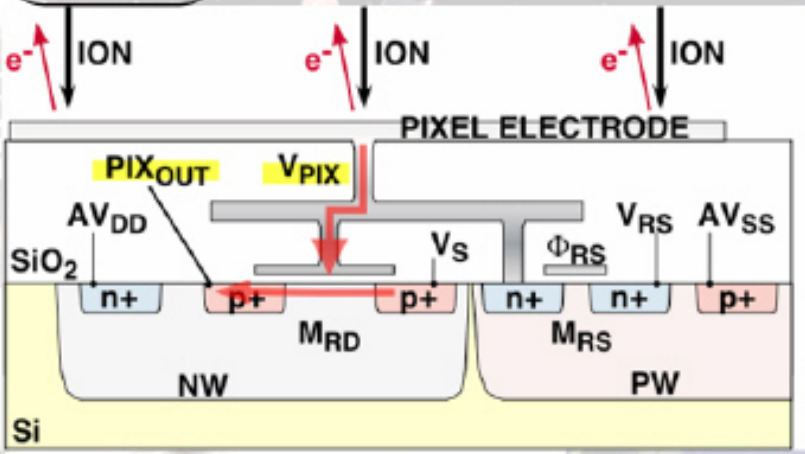

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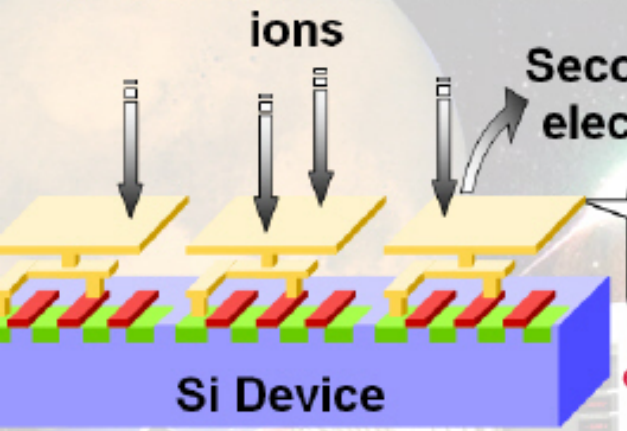

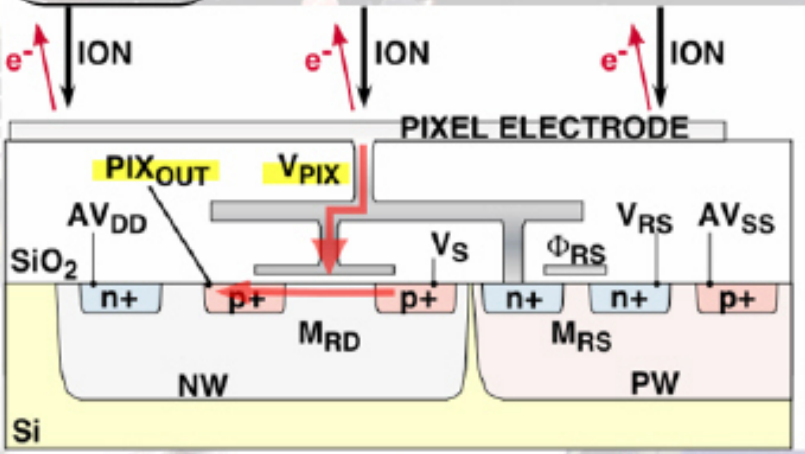

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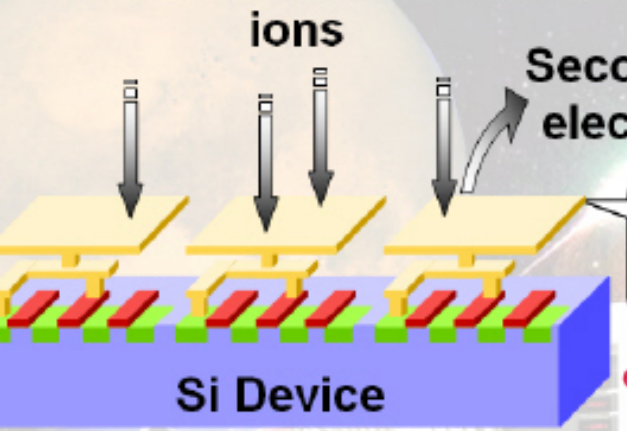

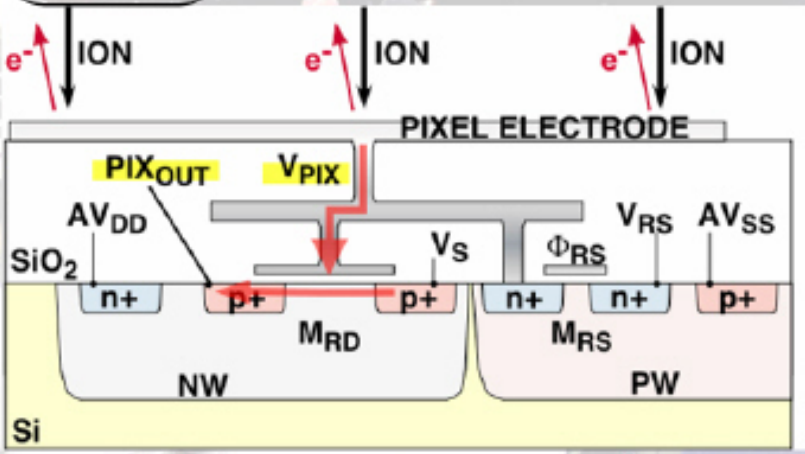

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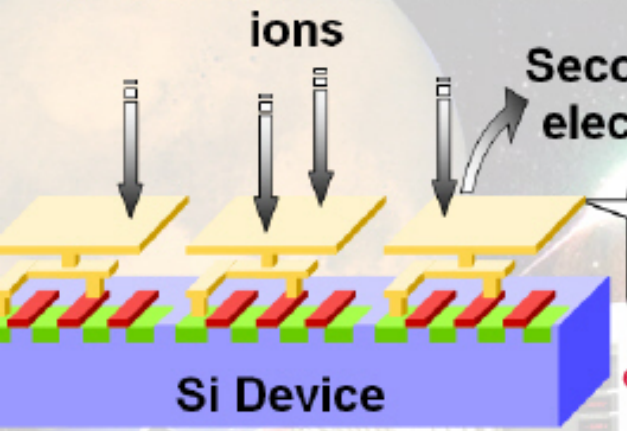

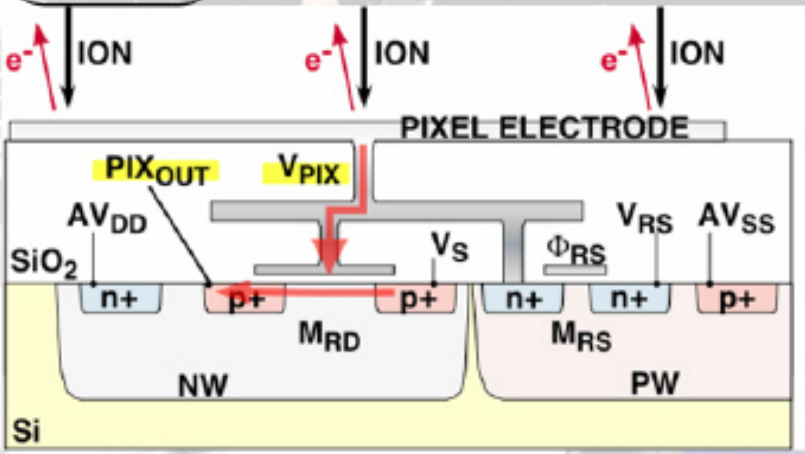

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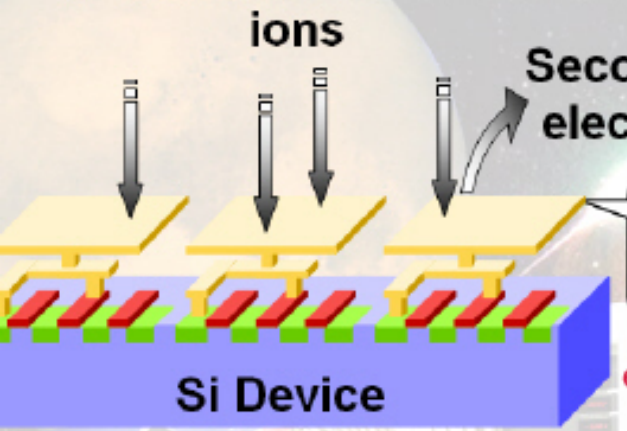

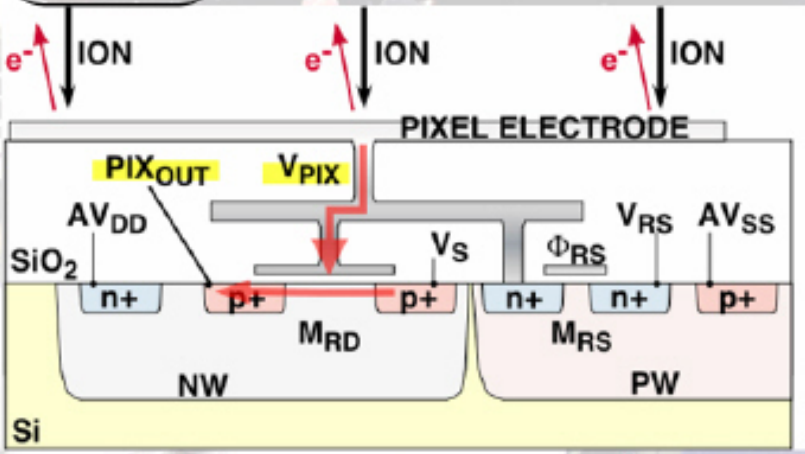

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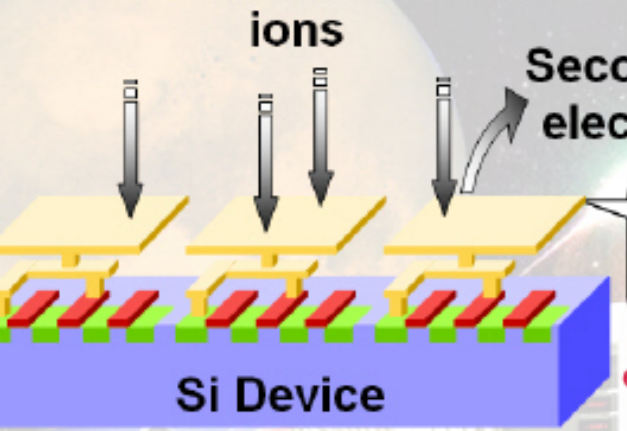

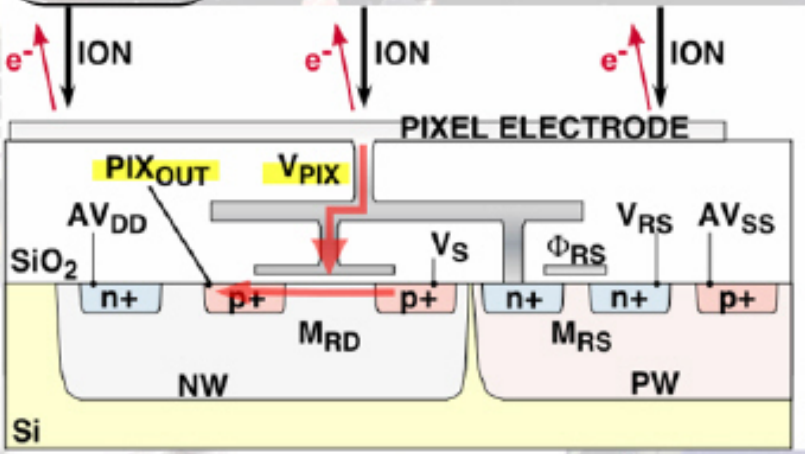

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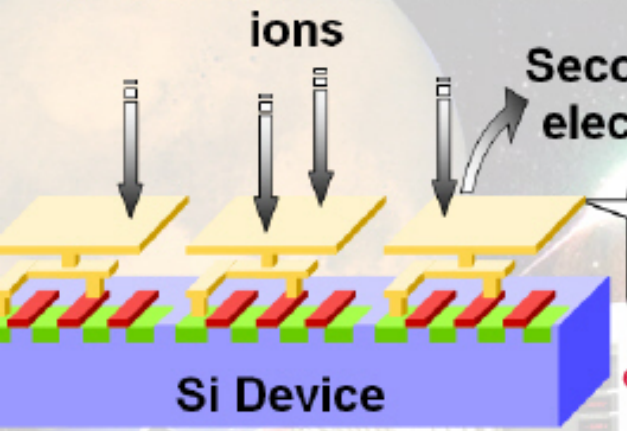

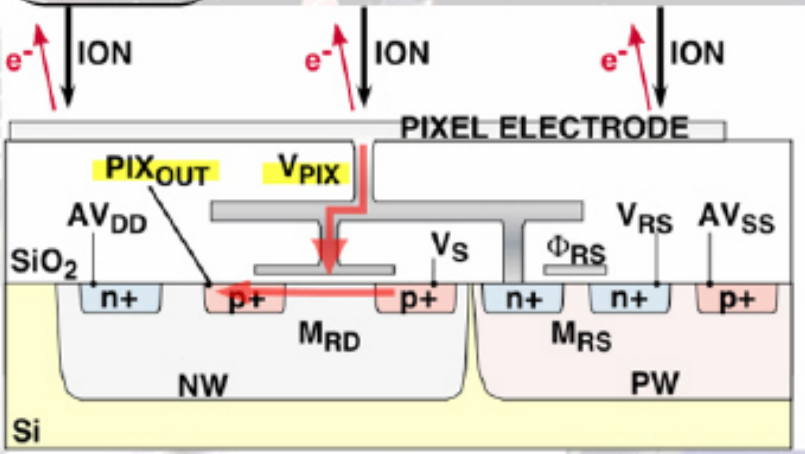

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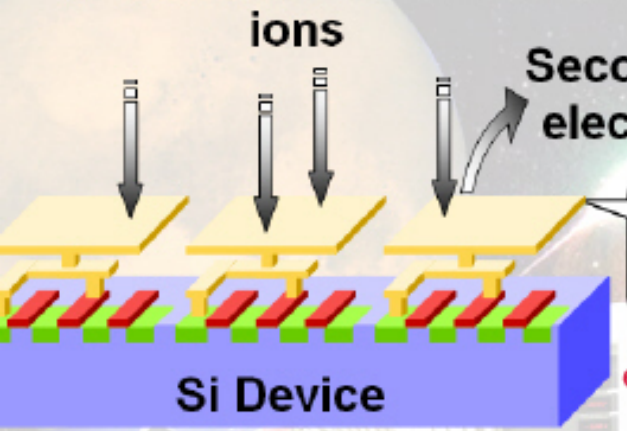

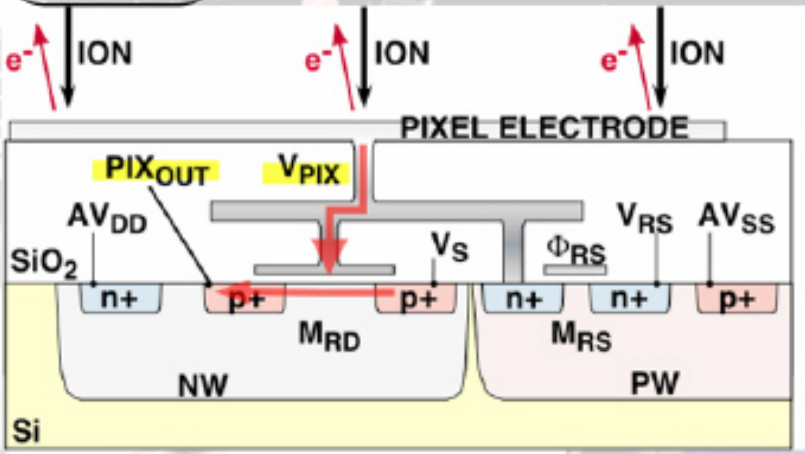

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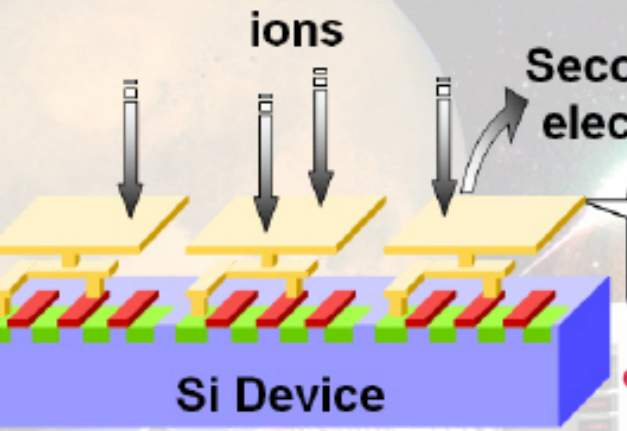
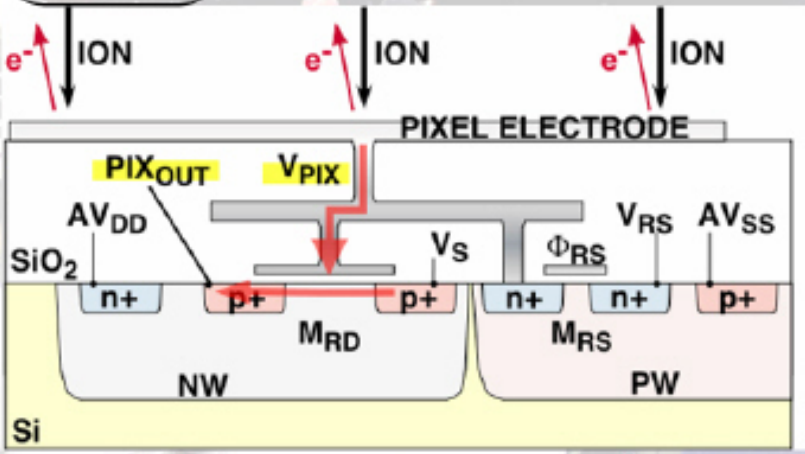
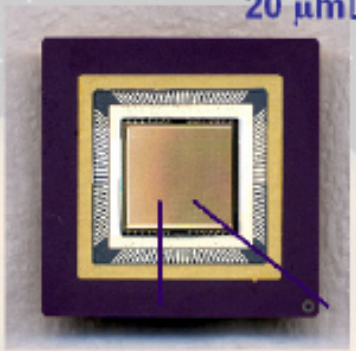


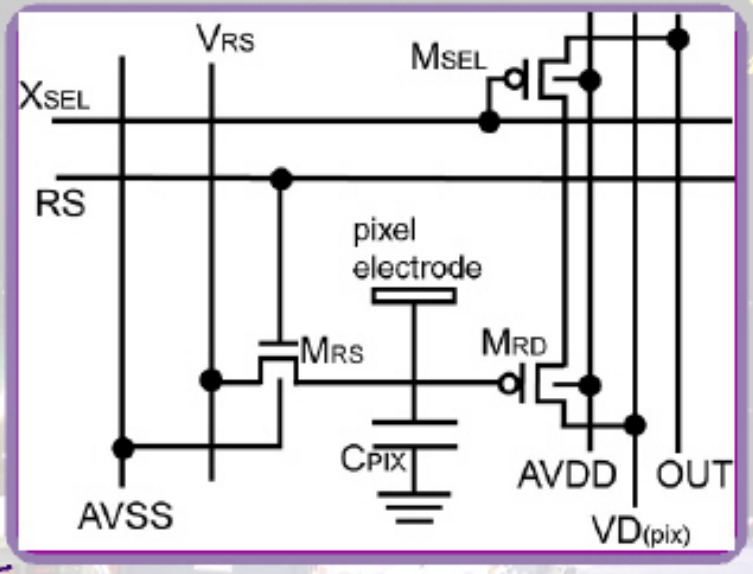


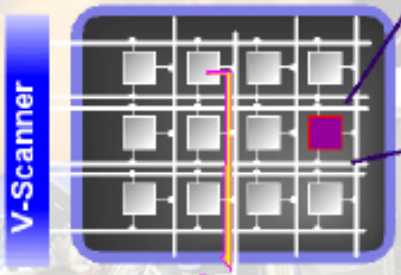
SCAPS

600 x 600 pixels
20 μm□/pixel



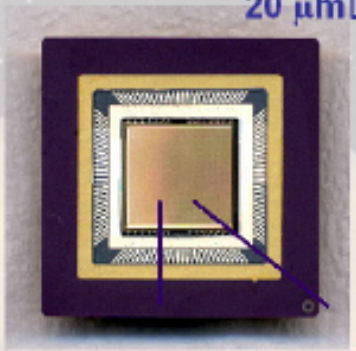
Pixel structure



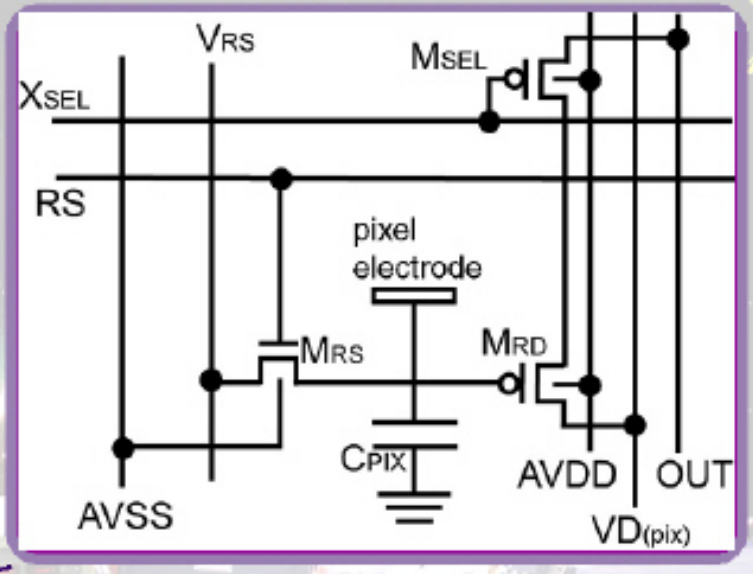


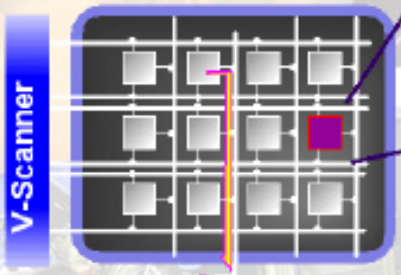
SCAPS

600 x 600 pixels
20 μm□/pixel



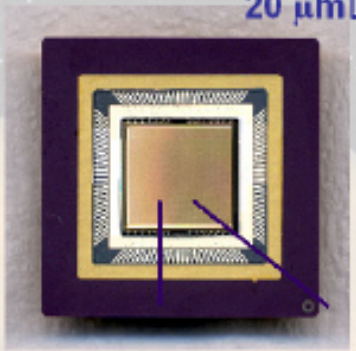
Pixel structure





SCAPS

600 x 600 pixels
20 μm□/pixel



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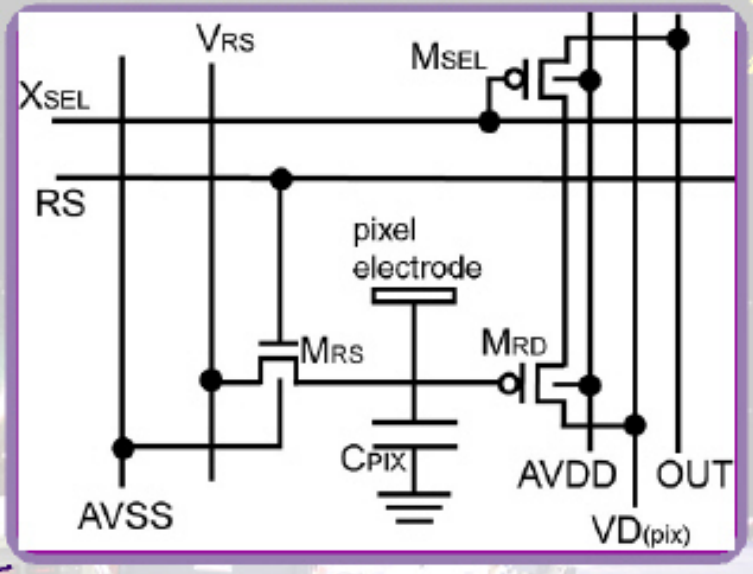

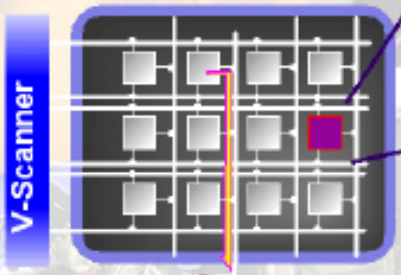

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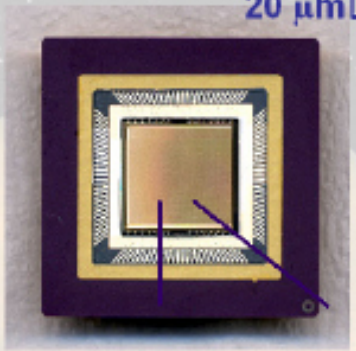

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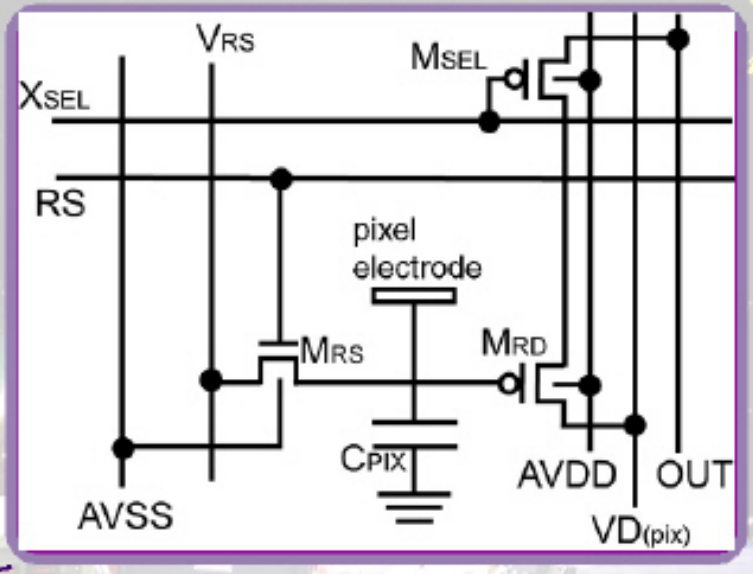

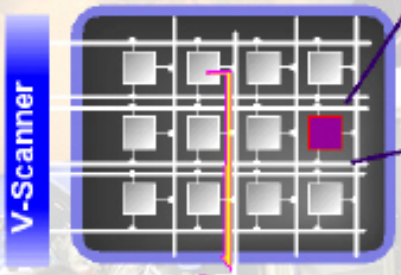

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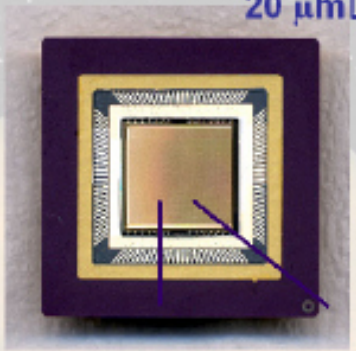

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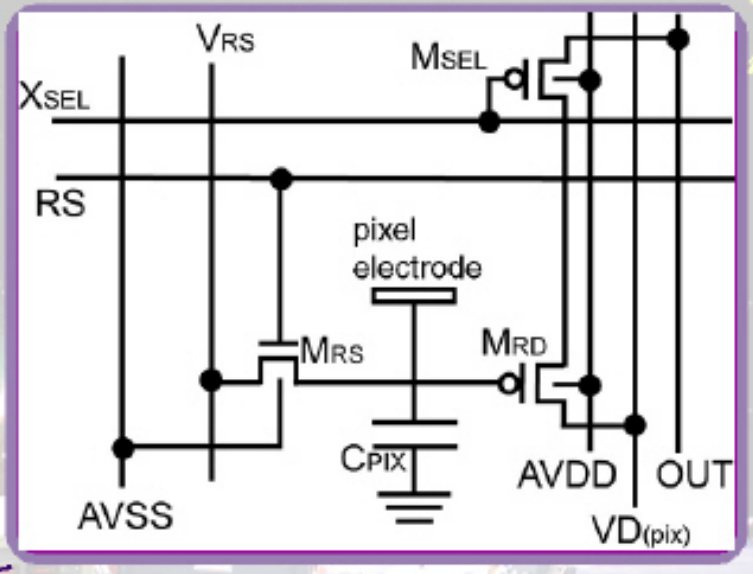
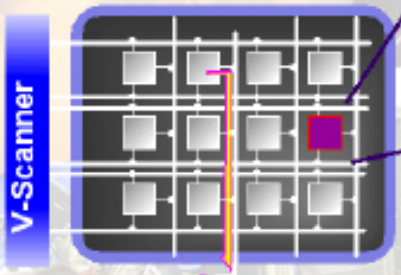
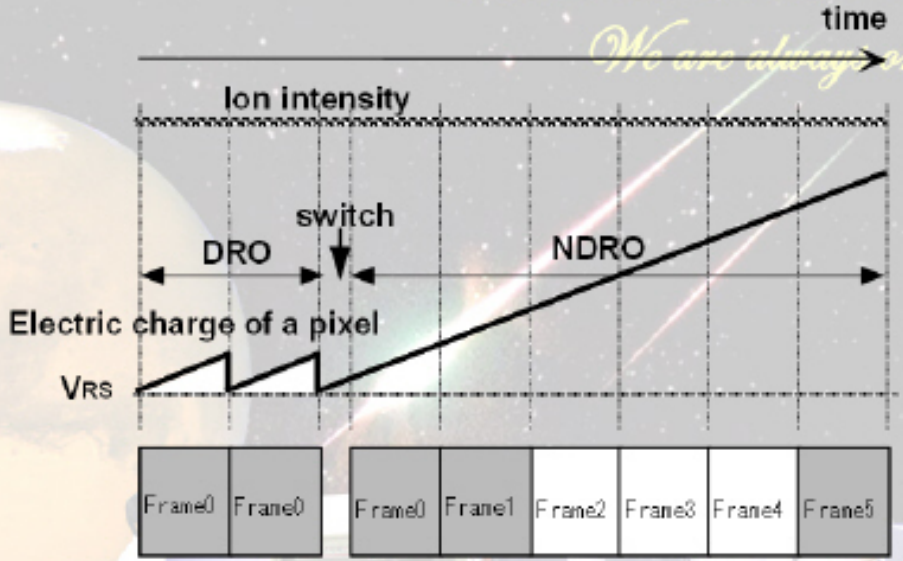


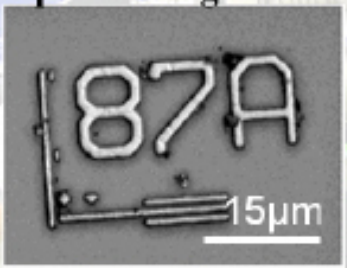


Non-destructive Readout (NDRO)

Cosmochemistry
We are always on the frontier.



Optical image



Non-destructive Readout (NDRO)

Nuclear Cosmochemistry
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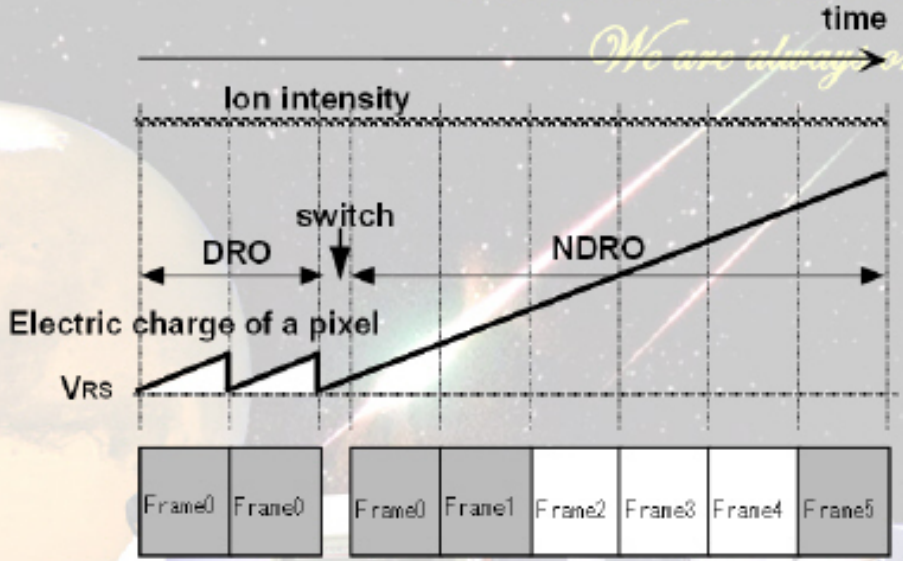

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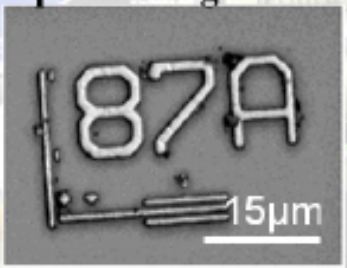
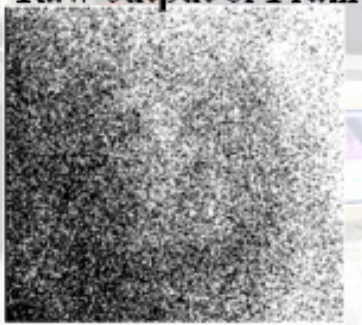


Raw output of Frame0



Non-destructive Readout (NDRO)

Nonlinear Cosmochemistry
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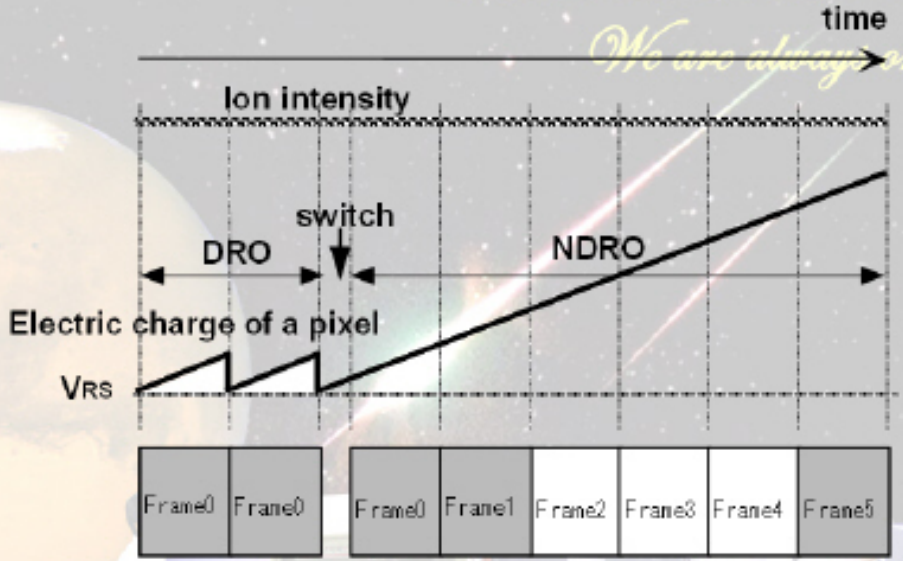
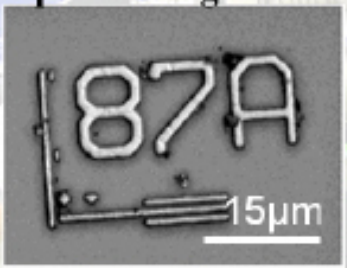
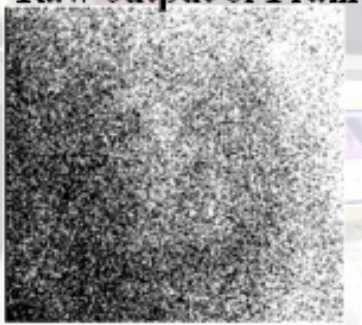


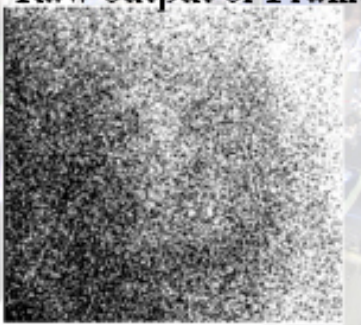
Optical image



Raw output of Frame0



Raw output of Frame1



Non-destructive Readout (NDRO)

Cosmochemistry
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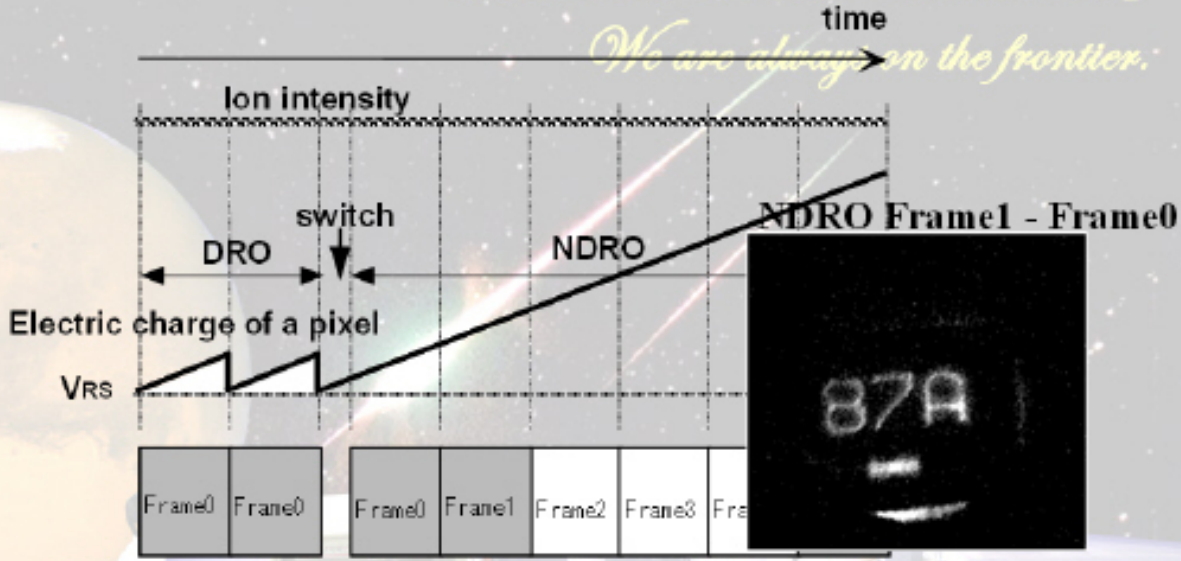
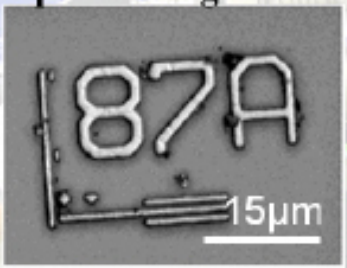
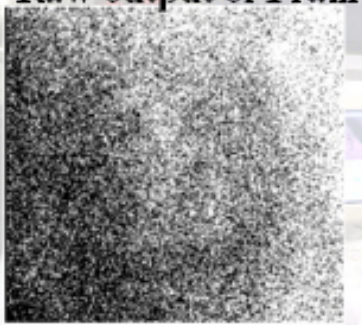


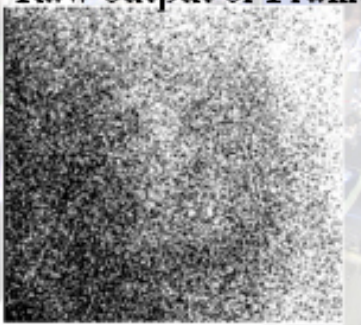
Optical image



Raw output of Frame0



Raw output of Frame1

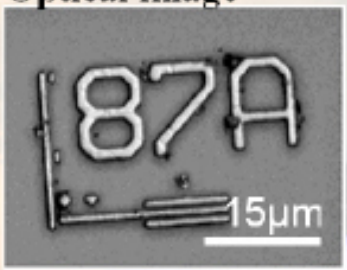


Reset frame correction

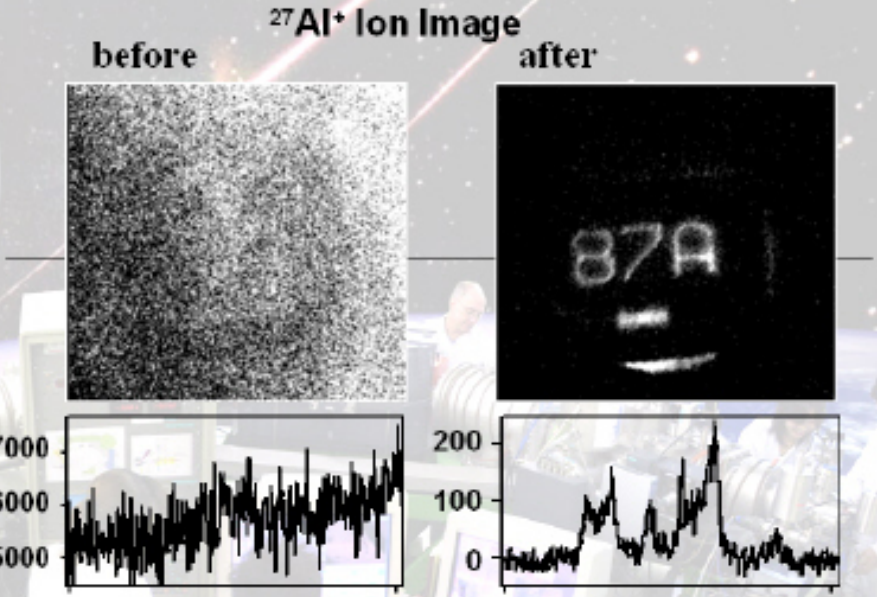
Hokudai Cosmochemistry

We are always on the frontier.

Optical image



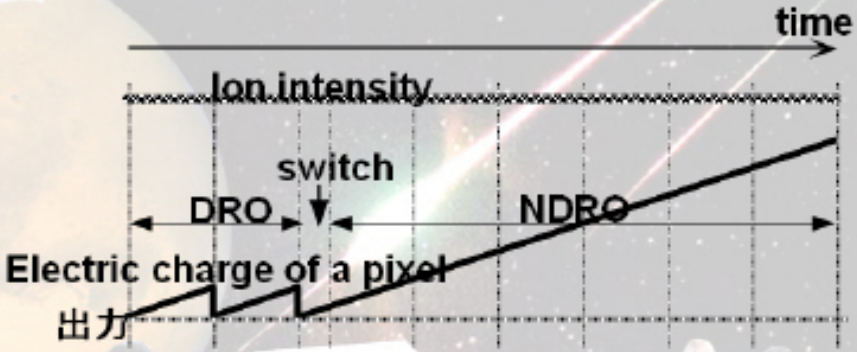
$^{27}Al^{+}$ Ion Image

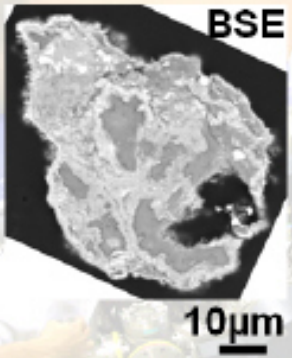


Reset Frame FPN: 600e-

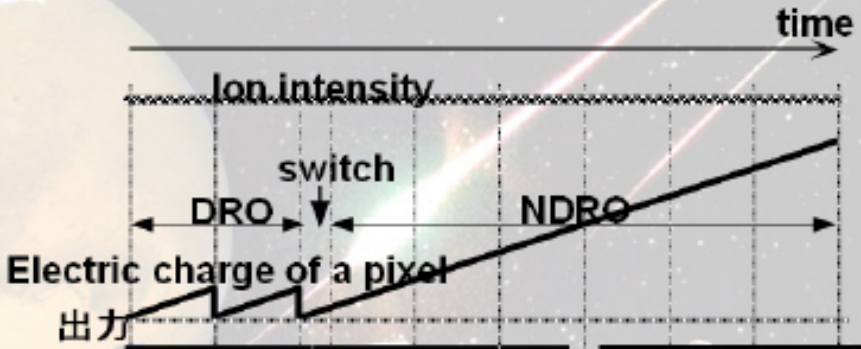
Reset noise : 25e-

Non-destructive Readout (NDRO)

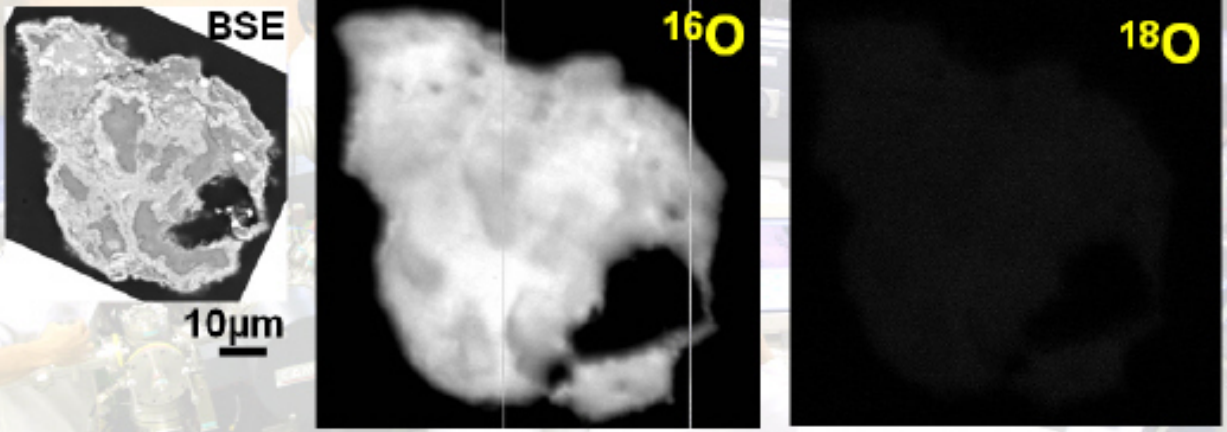




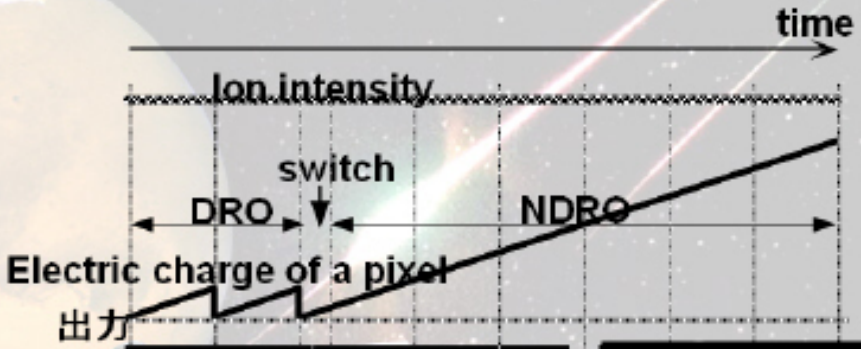
Non-destructive Readout (NDRO)



^{16}O~99.8%
^{18}O~ 0.2%



Non-destructive Readout (NDRO)



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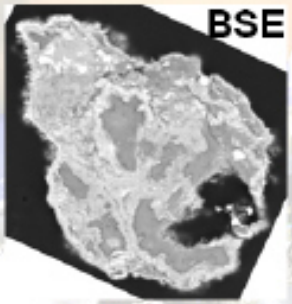
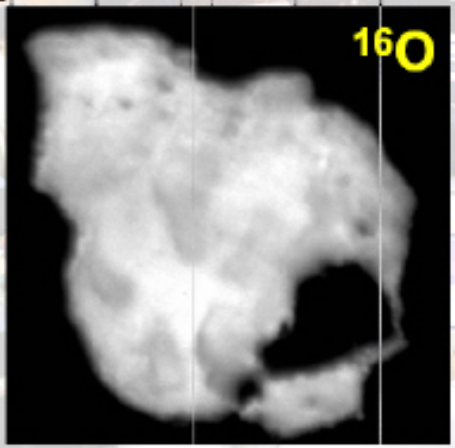
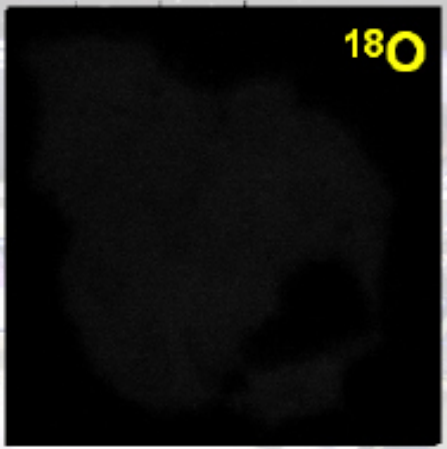
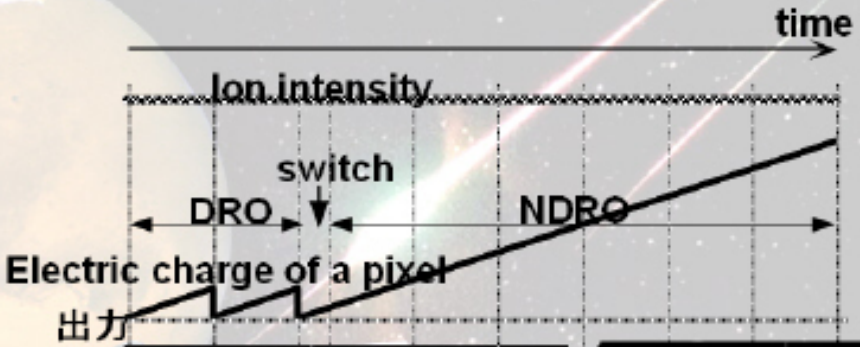






Non-destructive Readout (NDRO)

Isotope Cosmochemistry
We are always on the frontier.



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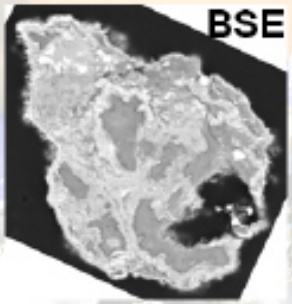
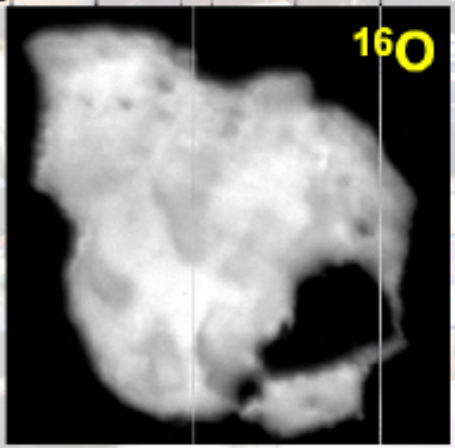
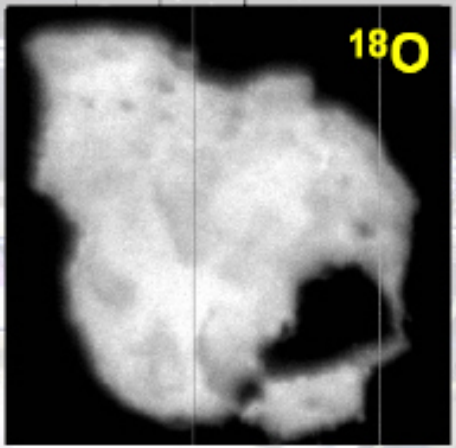
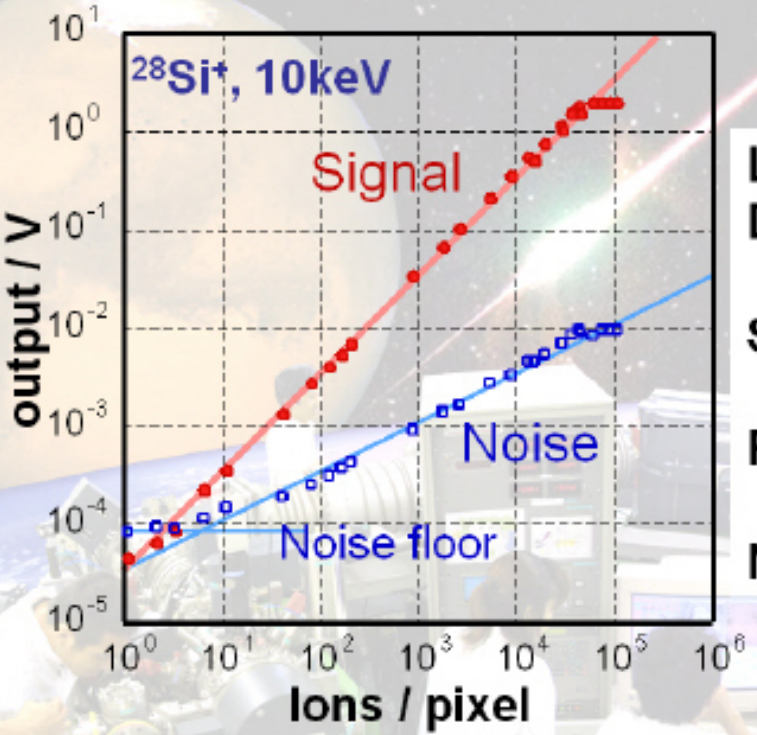






Linearity and Dynamic range

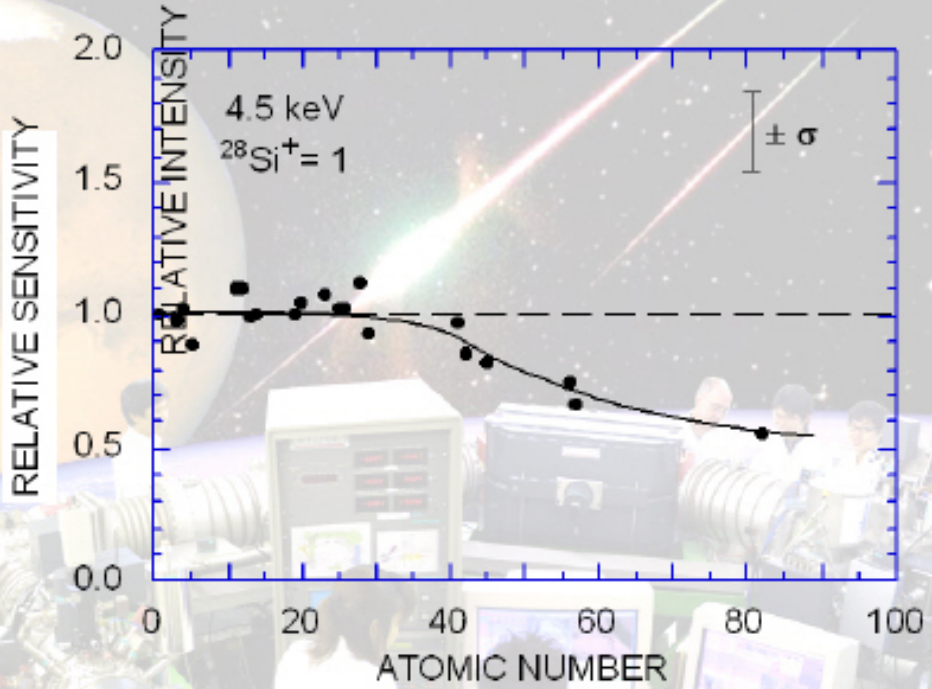
Kendal Cosmochemistry
We are always on the frontier.



Linear response
Dynamic Range
84 dB (~10^5)
Saturated ion numbers
5 x 10^4 ions@10keV
Readout noise
< 3 ions
Noise: statistic noise level

Relative sensitivity among ion species

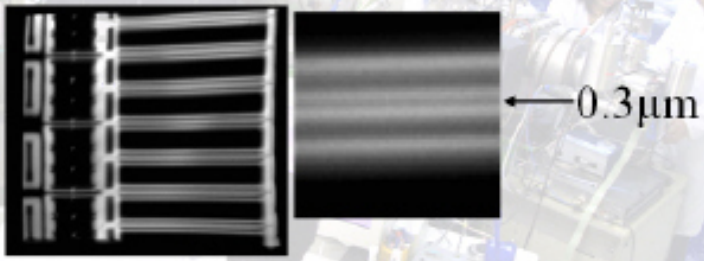
Isotope Cosmochemistry
We are always on the frontier.



SCAPS: a solution for ideal ion image detection

We are always on the frontier.

- Sensitive for ions from single ion
- 600 × 600 pixels (20 x 20 μm/pixel)
- Small dead space (high-aperture-ratio pixel design; 88%)
- NO dead time
- Monitoring of accumulating state for ions
- Small readout noise
- Wide dynamic range
- constant sensitivity for all ions
- Robustness
- Slow readout



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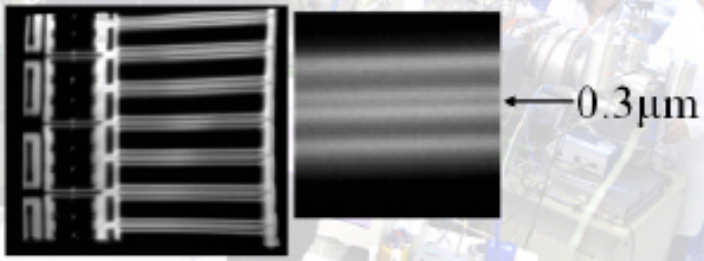

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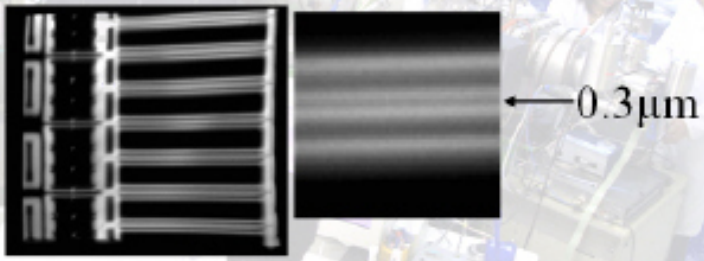

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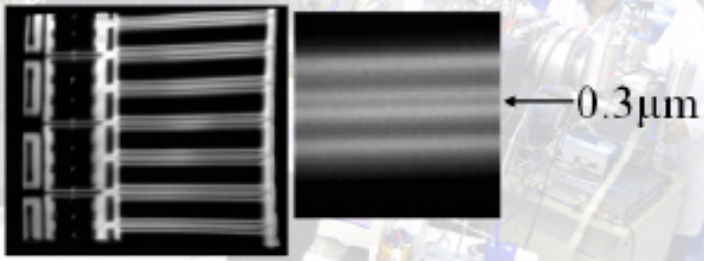

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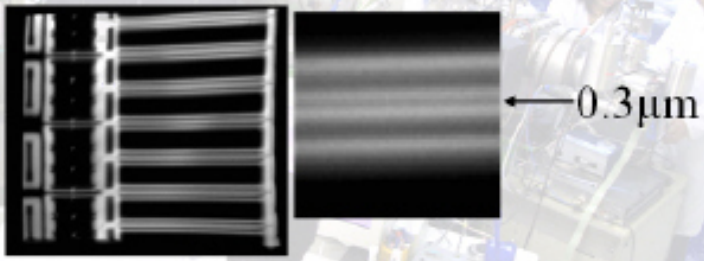

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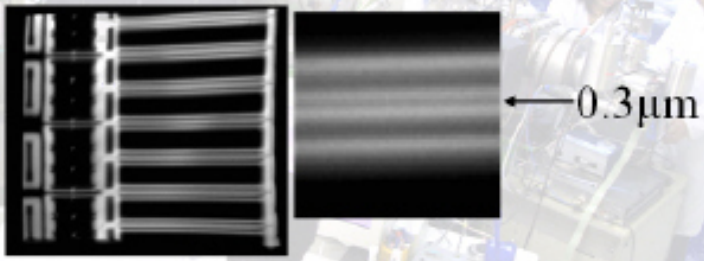

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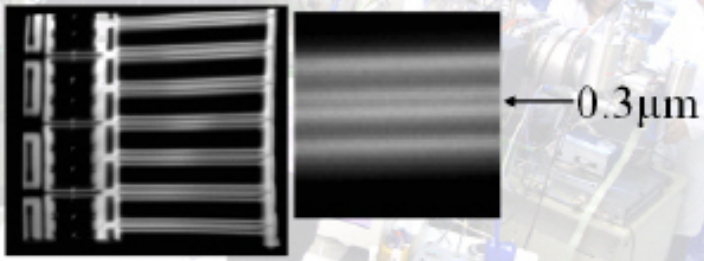

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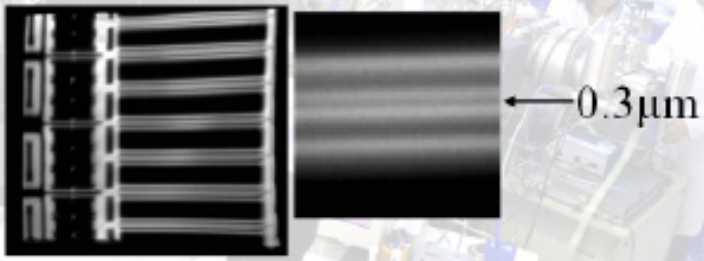
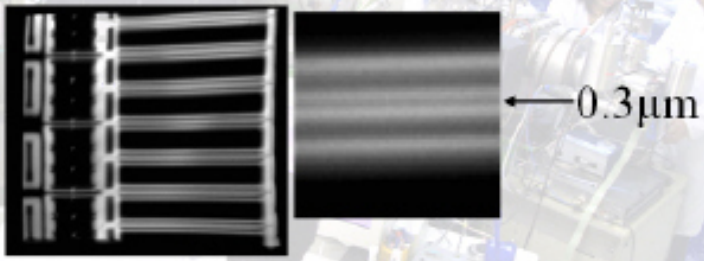
- Small dead space (high-aperture-ratio pixel design; 88%)
- NO dead time
- Monitoring of accumulating state for ions
- Small readout noise
- Wide dynamic range
- constant sensitivity for all ions
- Robustness
- Slow readout



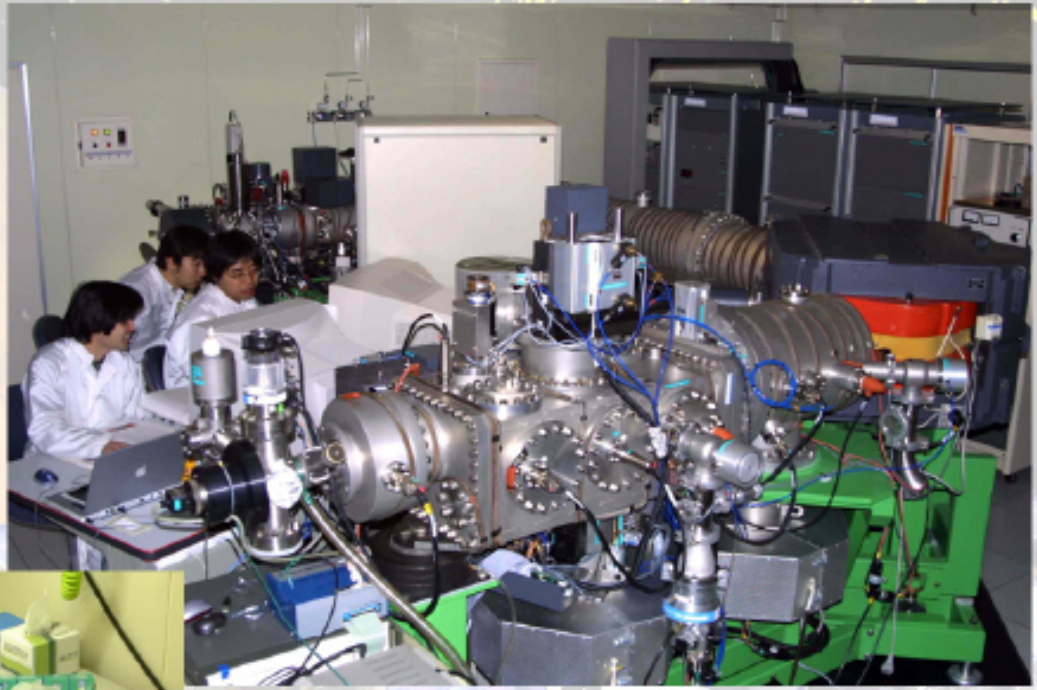
SCAPS: a solution for ideal ion image detection

We are always on the frontier.

- Sensitive for ions from single ion
- 600 × 600 pixels (20 x 20 μm/pixel)
- Small dead space (high-aperture-ratio pixel design; 88%)
- NO dead time
- Monitoring of accumulating state for ions
- Small readout noise
- Wide dynamic range
- constant sensitivity for all ions
- Robustness
- Slow readout



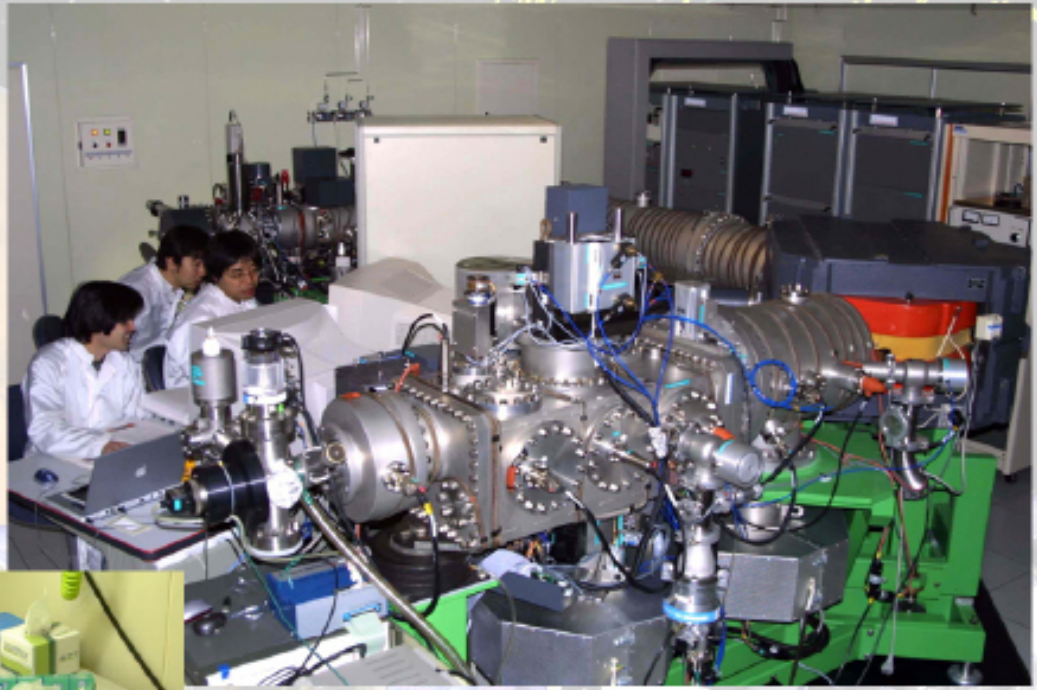
Isotope microscope system



Cameca ims 1270

SCAPS

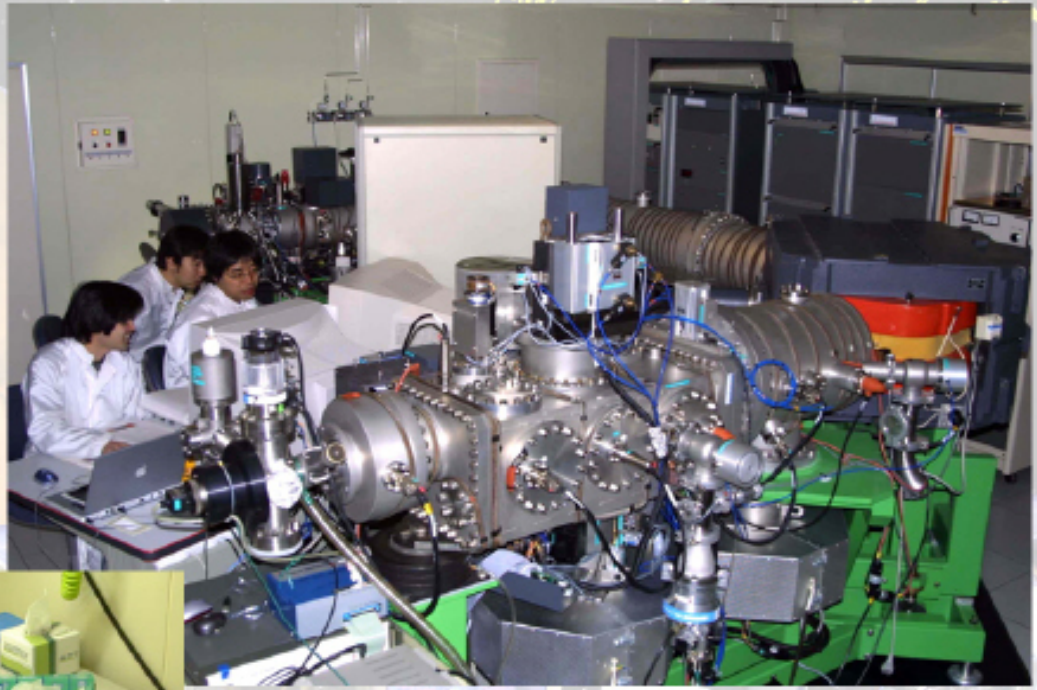
Isotope microscope system



Cameca ims 1270

SCAPS

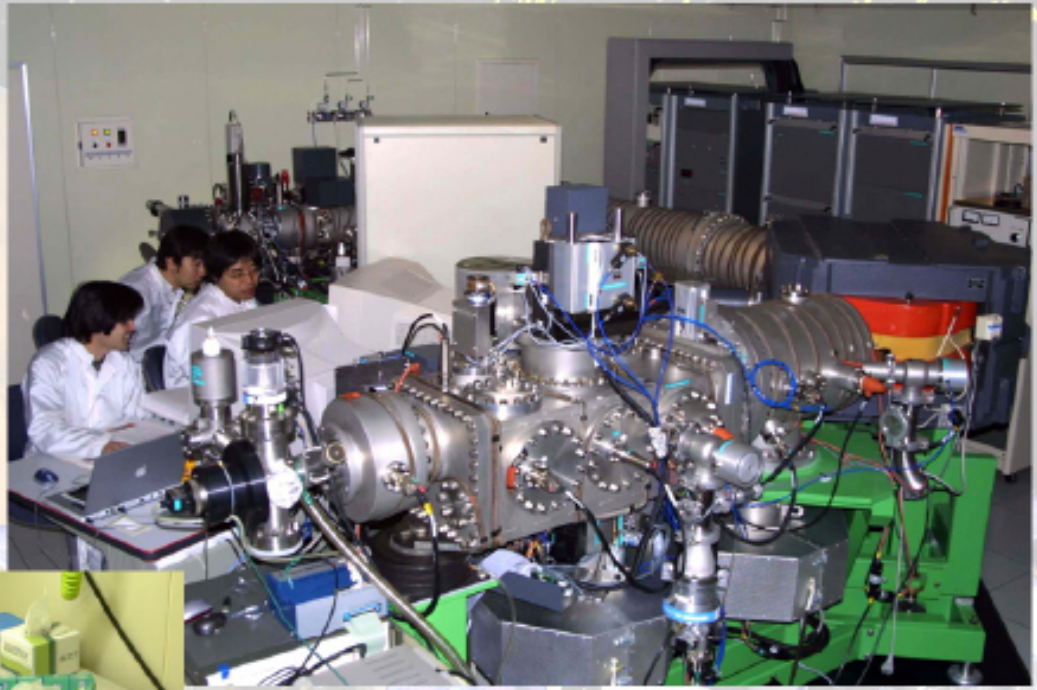
Isotope microscope system



Cameca ims 1270

SCAPS

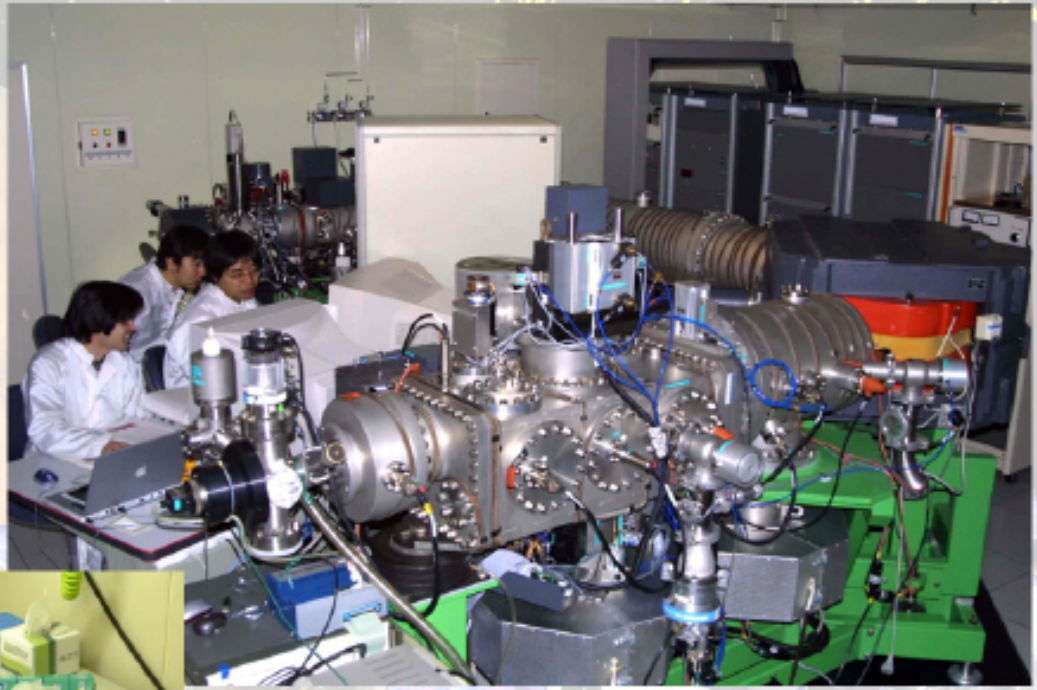
Isotope microscope system



Cameca ims 1270

SCAPS

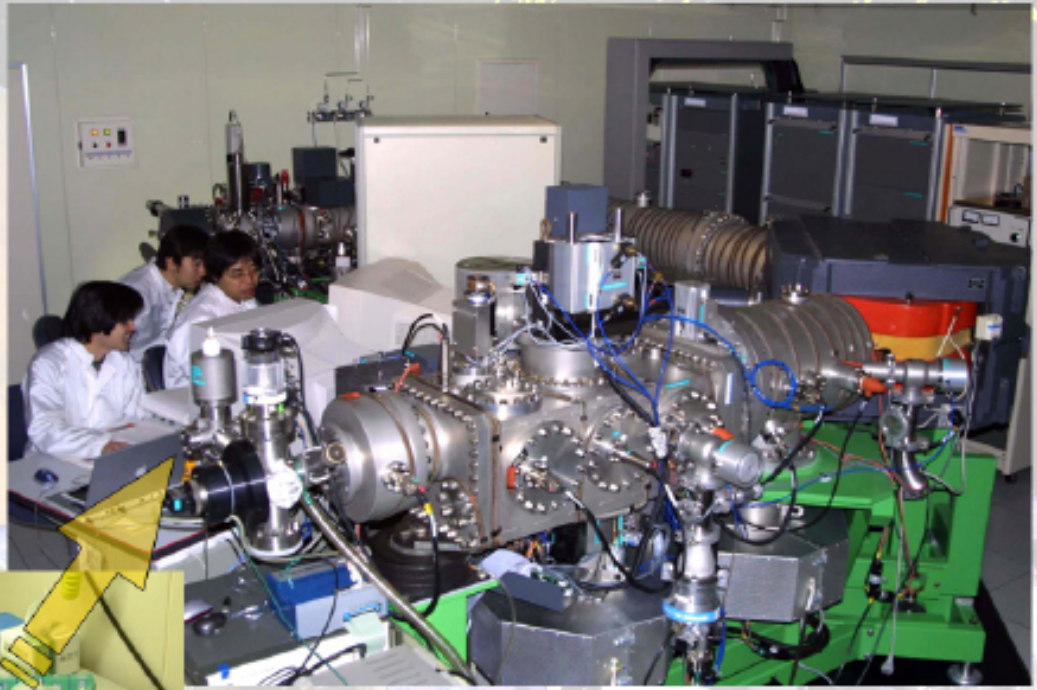
Isotope microscope system



Cameca ims 1270

SCAPS

Isotope microscope system



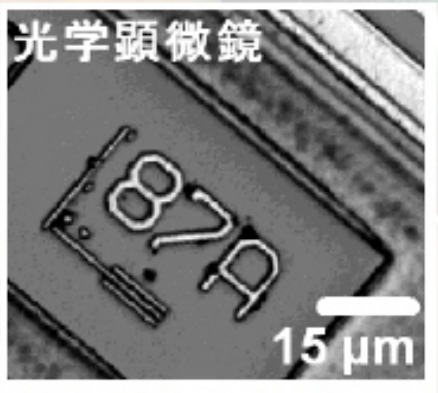
Cameca ims 1270

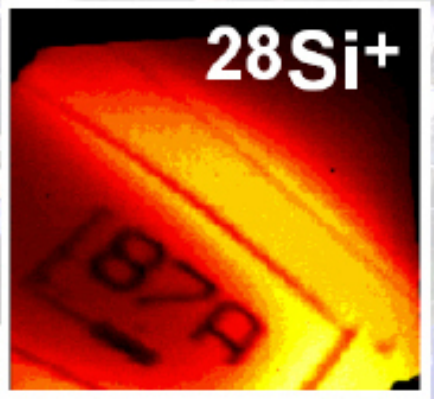
SCAPS

Example of isotope image

Hokudai Cosmochemistry

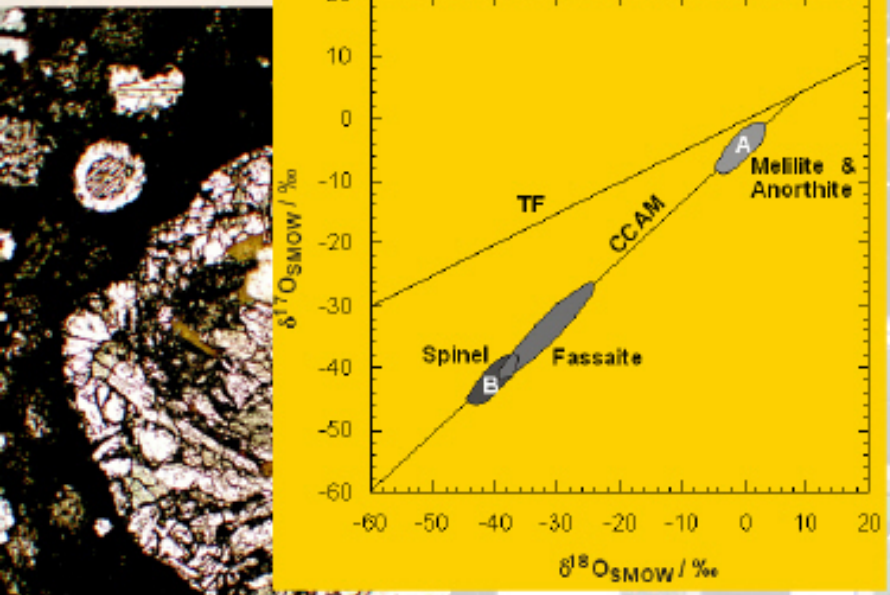
We are always on the frontier.





O Isotope distribution in CAI

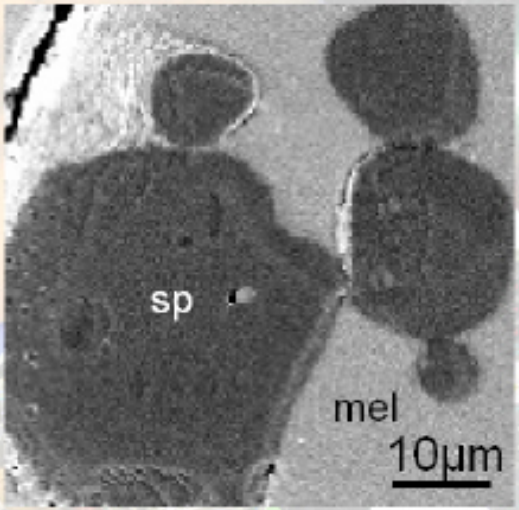
Isotopic Cosmochemistry
We are always on the frontier.

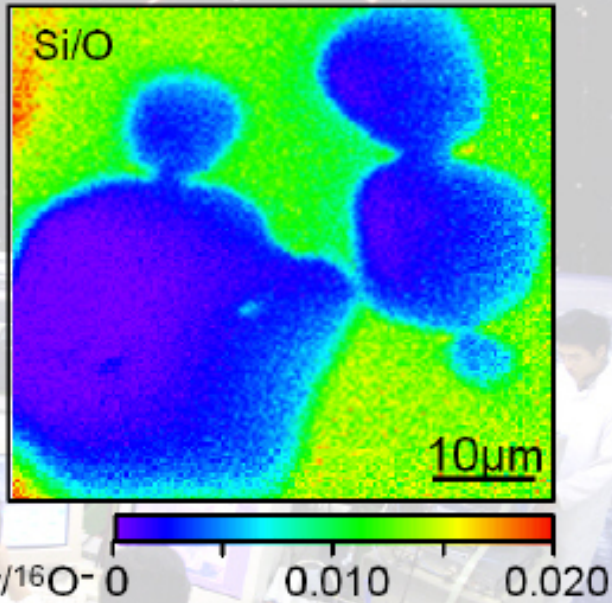


- Inter-mineral : 40‰
- Intra-minaral : homogeneous
  - <0.1‰

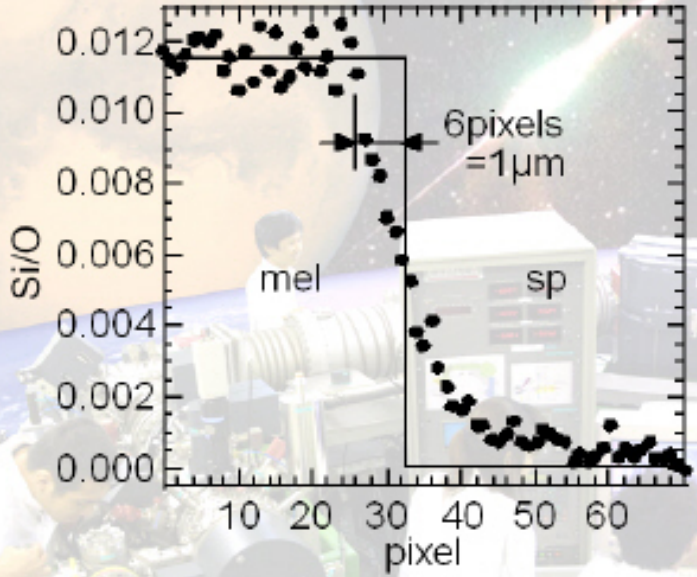
BSE vs secondary ion image

Meteoritical Cosmochemistry
We are always on the frontier.





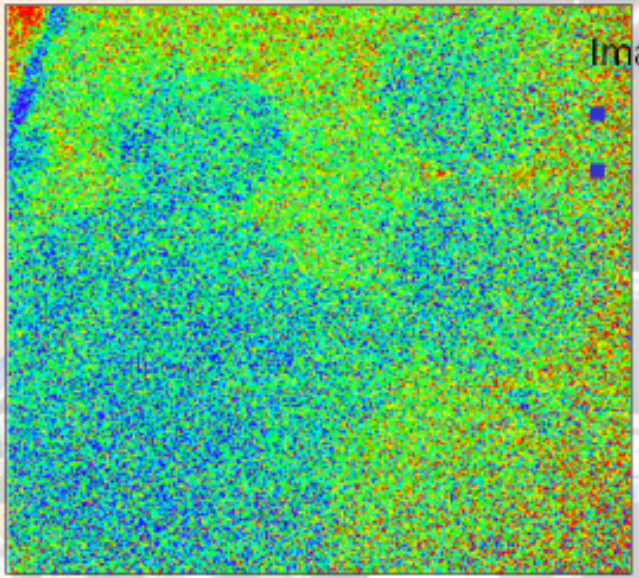
Spatial resolution



- 1μm
- Restricted by aberration of the ion optics

O Isotope image

- (^{16}O, ^{17}O, ^{18}O)= (99.763, 0.0372, 0.1995)
- Not sufficient intensity per pixel

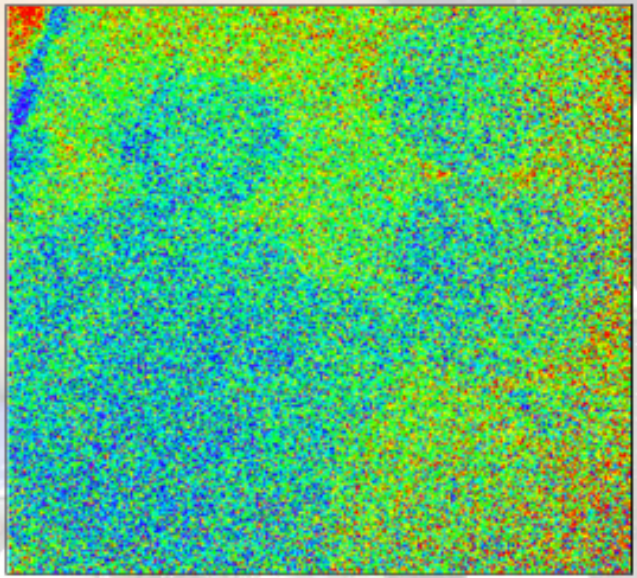


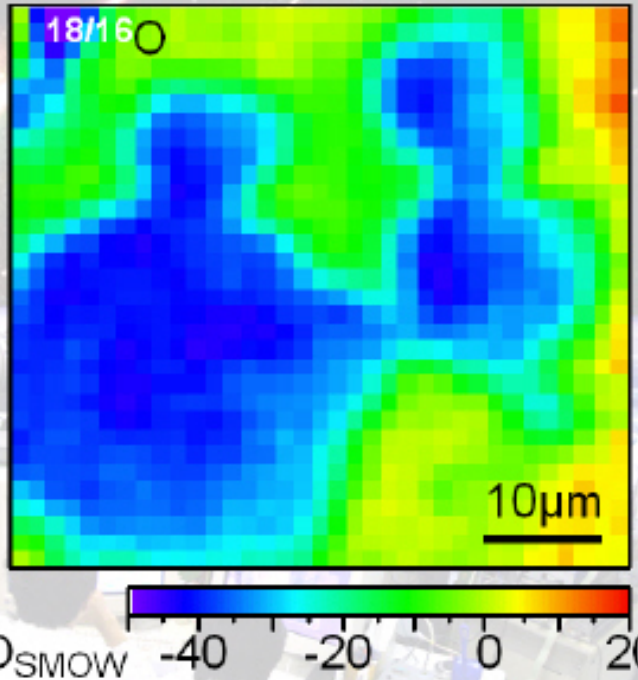
$^{18}O/^{16}O$

Image processing

- 9 x 9 binning
- Smoothing by moving-average

Effect of image processing





For ^{17}O imaging

Hokudai Cosmochemistry

We are always on the frontier.

- High mass resolution
  - $M/\Delta M = 5000$

Suppressing of ^{16}OH interference

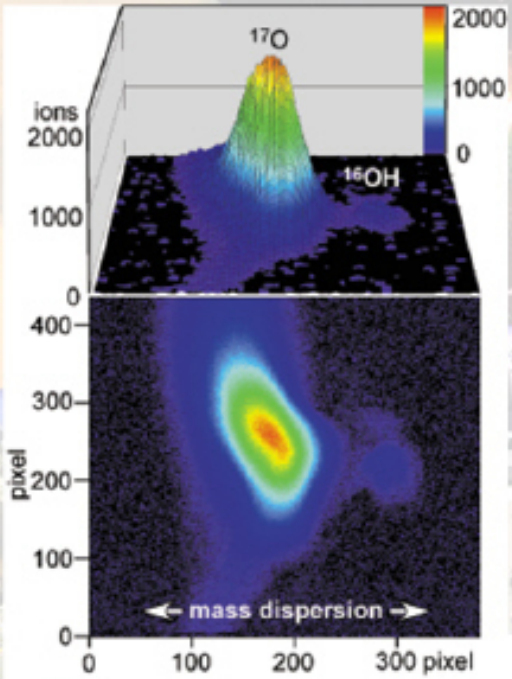
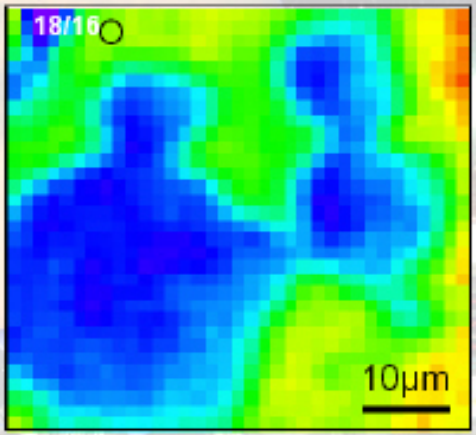


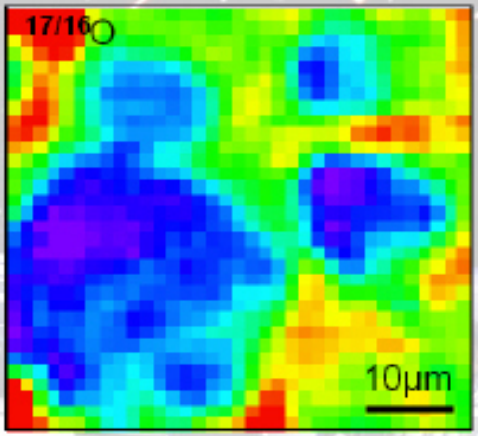
Fig. 2

$^{18}O/^{16}O$ vs $^{17}O/^{16}O$

Hokudai Cosmochemistry

We are always on the frontier.

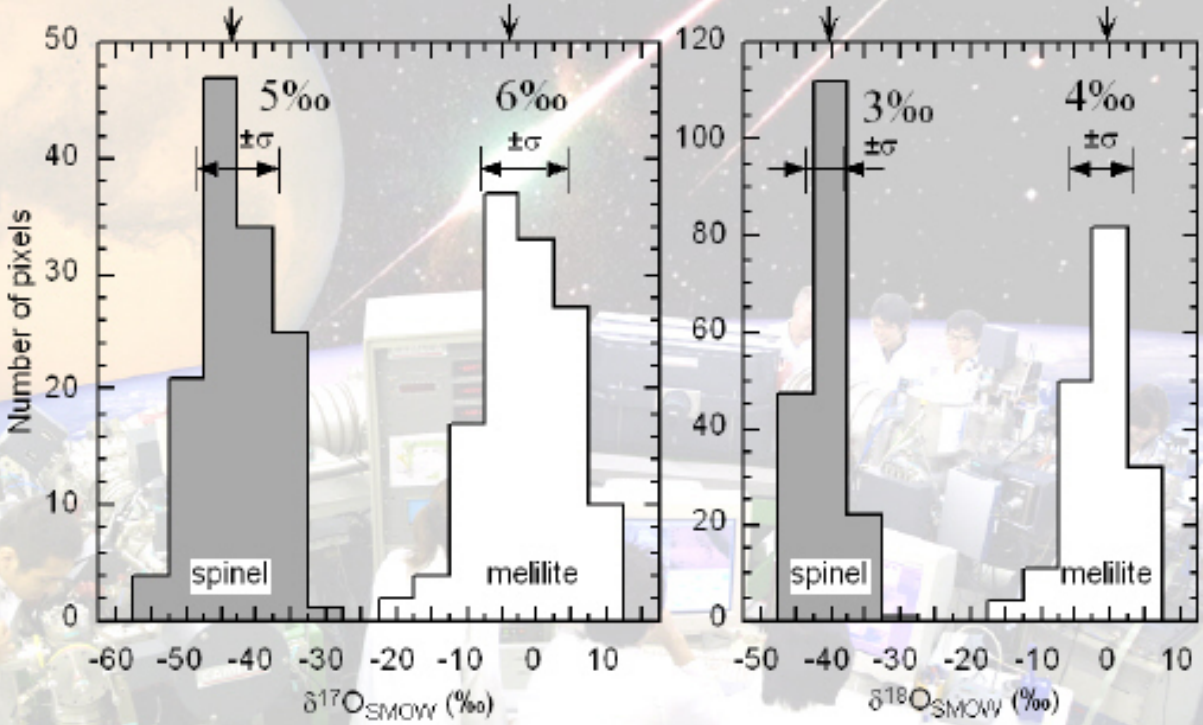




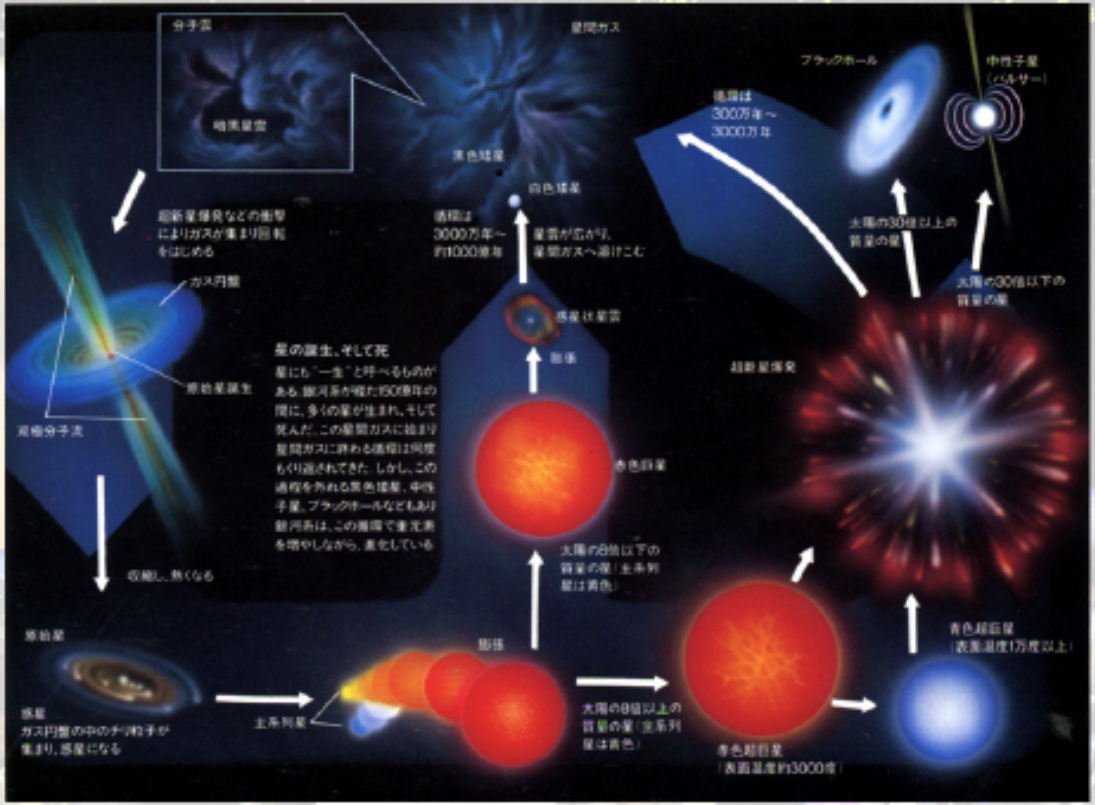
Evaluation by conventional results

Hokudai Cosmochemistry

We are always on the frontier.

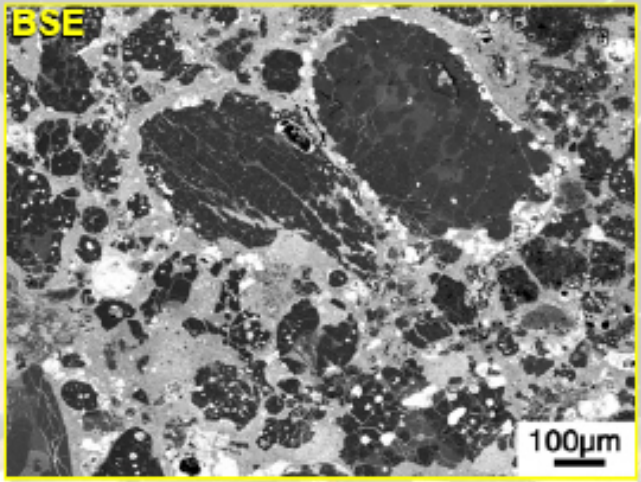


Application I: Survey of presolar grains



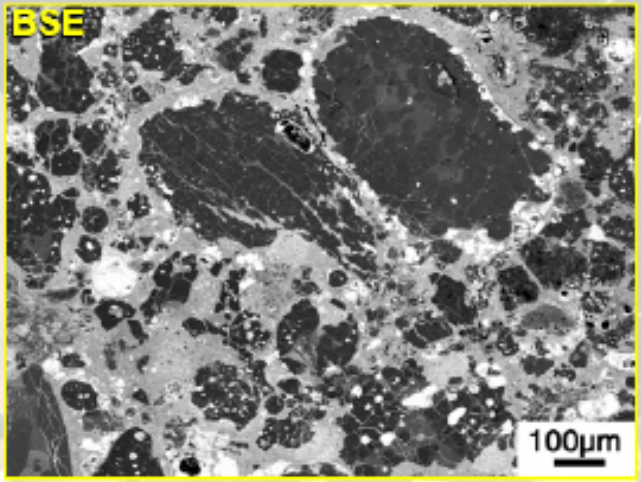
Application I: Survey of presolar grains

NanoSIMS Cosmochemistry
We are always on the frontier.



Application I: Survey of presolar grains

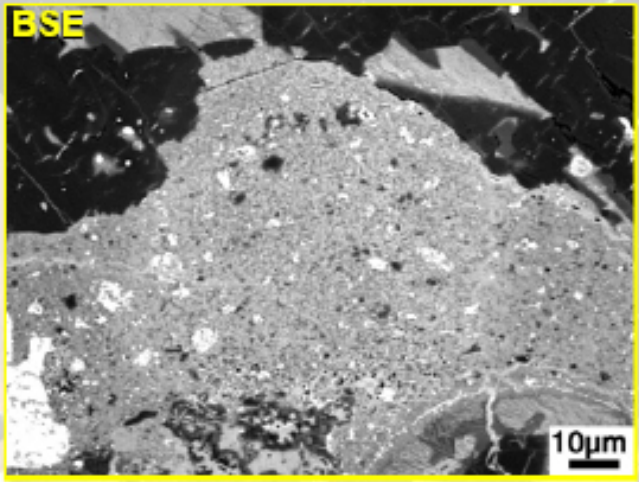
NanoSIMS Cosmochemistry
We are always on the frontier.



Adelaide (ungrouped)

Application I: Survey of presolar grains

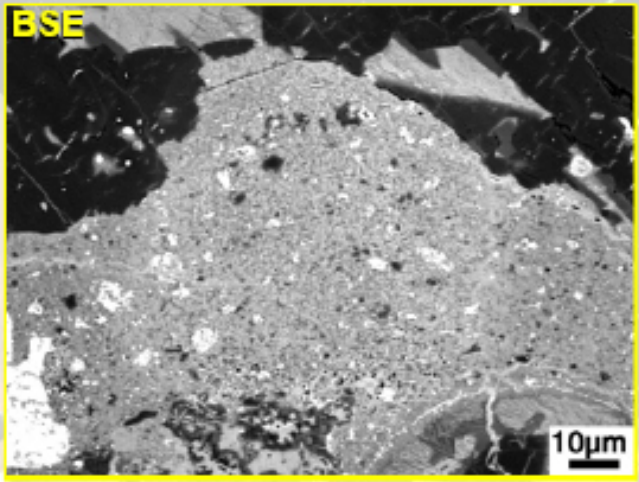
NASA Cosmochemistry
We are always on the frontier.



Adelaide (ungrouped)

Application I: Survey of presolar grains

NASA Cosmochemistry
We are always on the frontier.



Adelaide (ungrouped)

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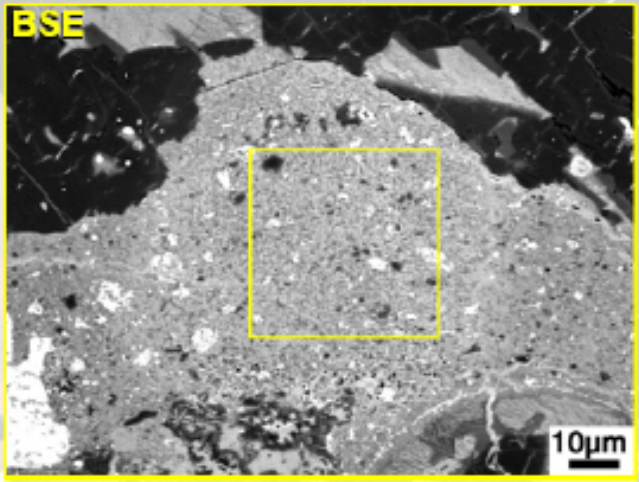
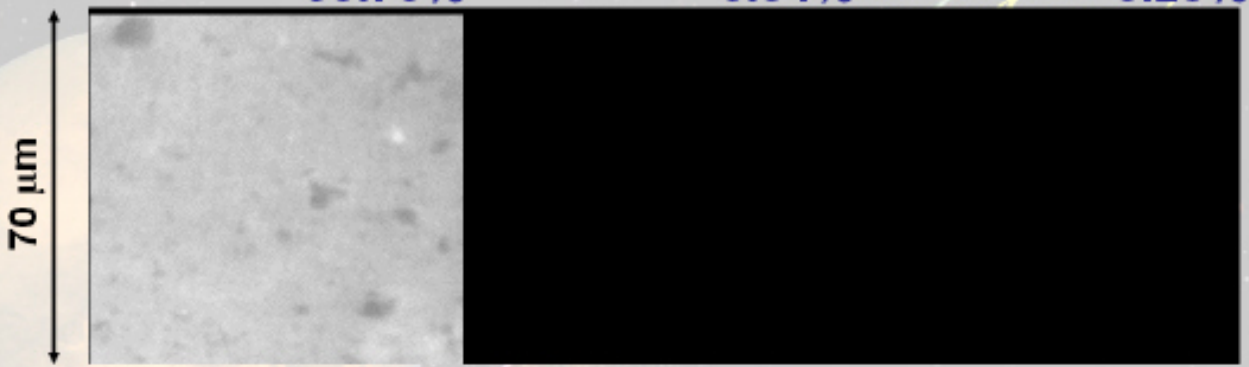
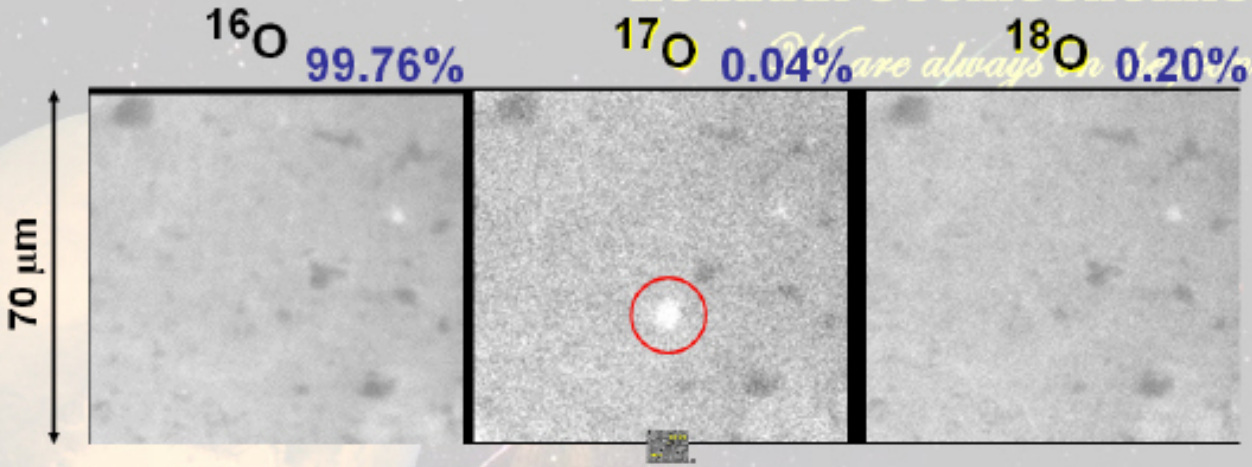


Adelaide (ungrouped)

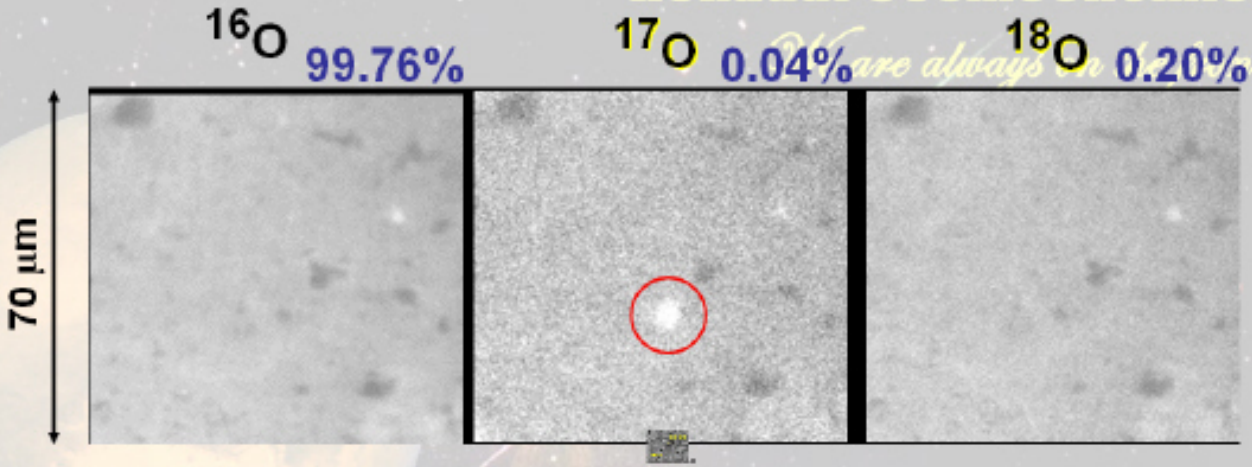
Application I: Survey of presolar grains



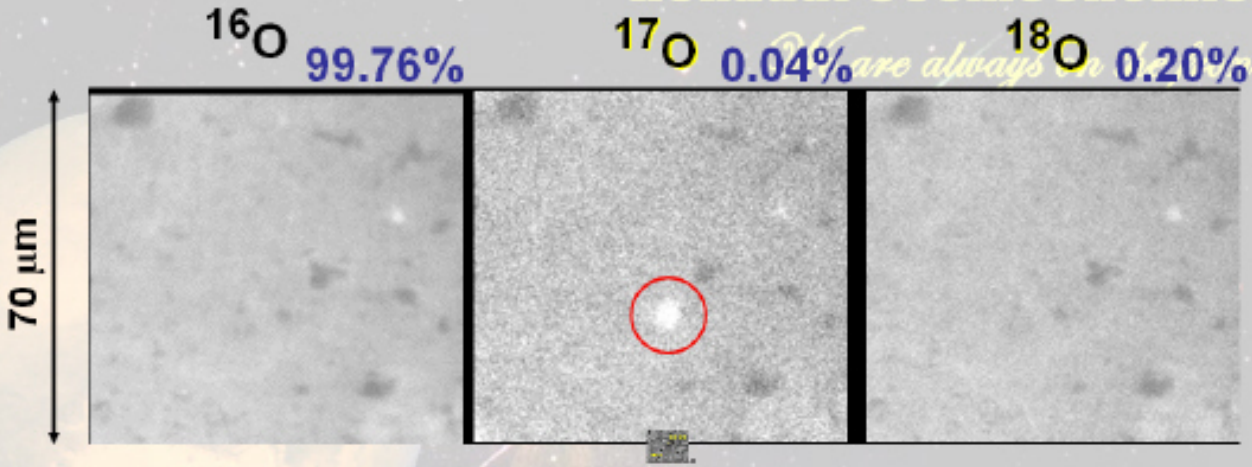
Application I: Survey of presolar grains



Application I: Survey of presolar grains



Application I: Survey of presolar grains



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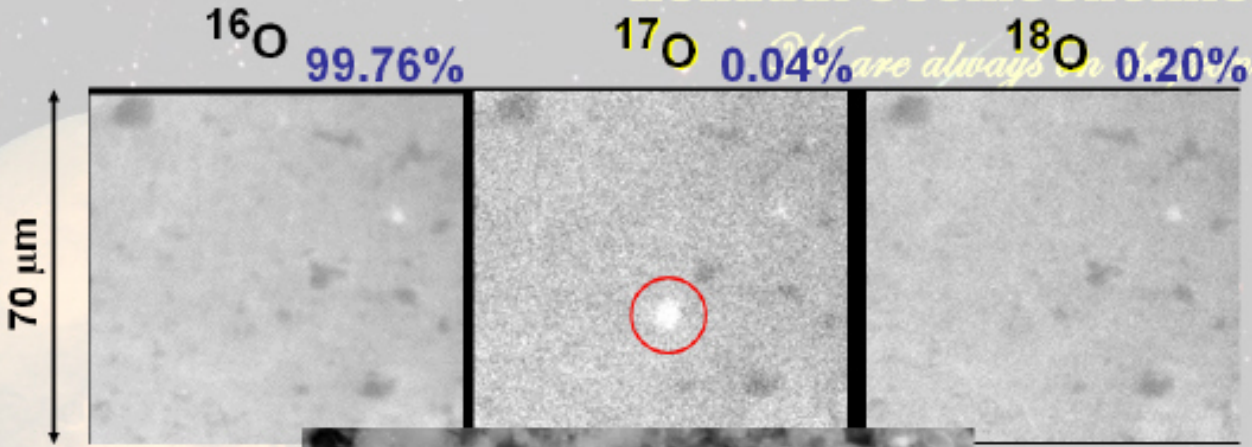
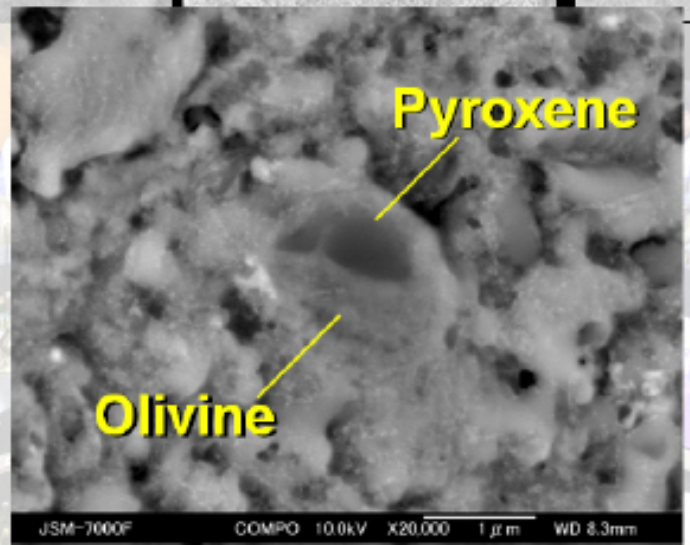
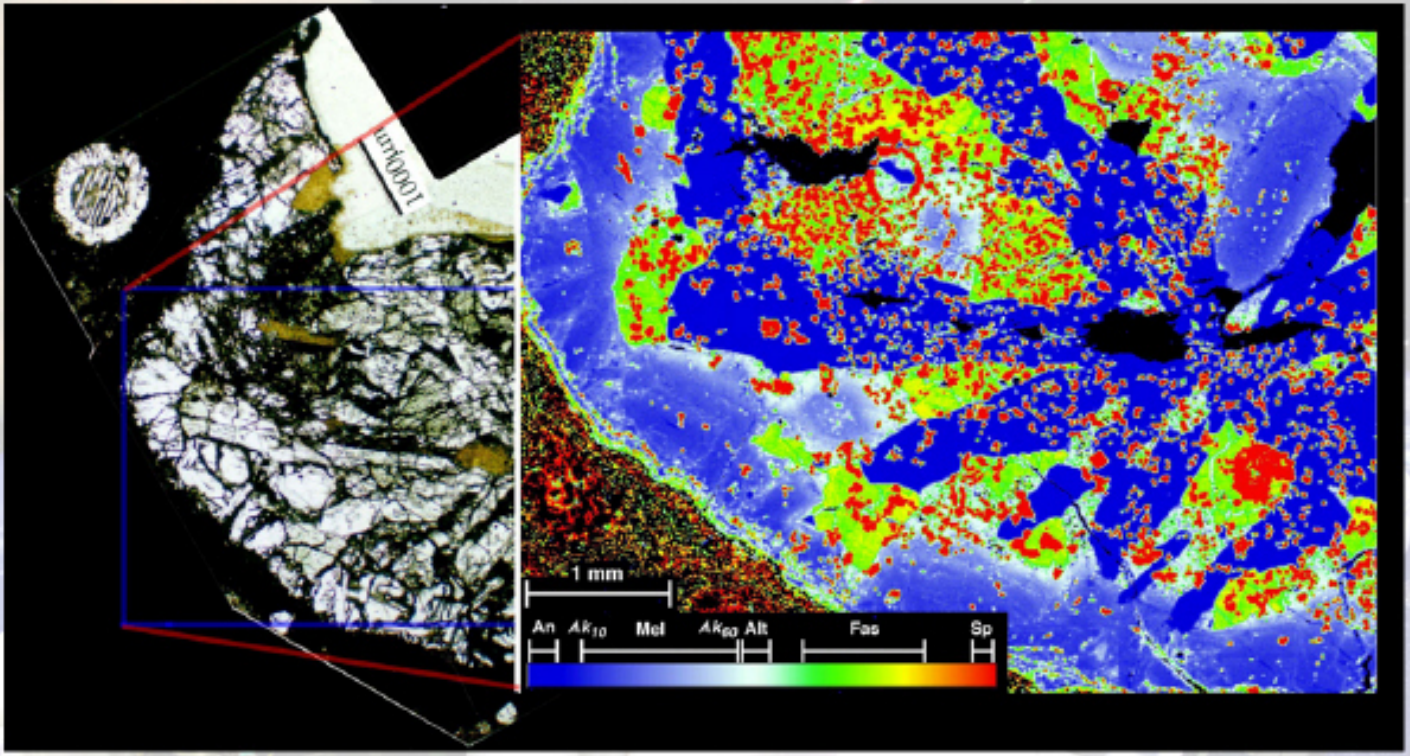




1μm

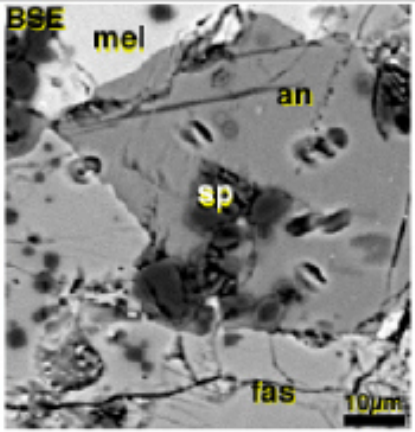
Application II: Oxygen isotopic variation in the early solar system

Nuclear Cosmochemistry
We are always on the frontier.



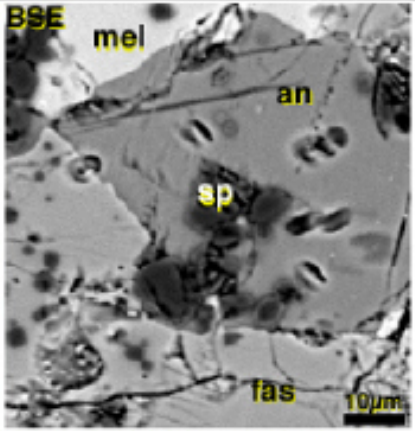
Application II: Oxygen isotopic variation in the early solar system

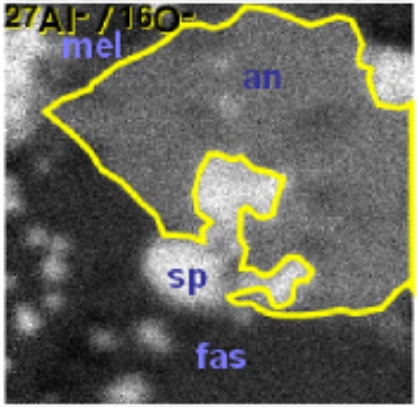
Nuclear Cosmochemistry
We are always on the frontier.



Application II: Oxygen isotopic variation in the early solar system

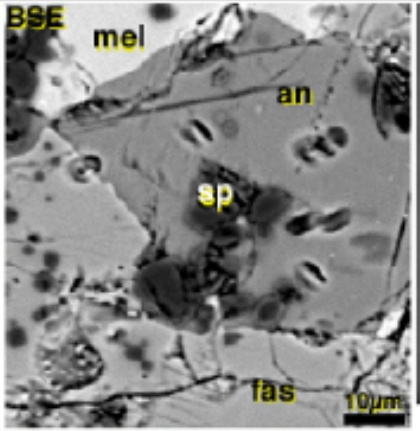
Nuclear Cosmochemistry
We are always on the frontier.

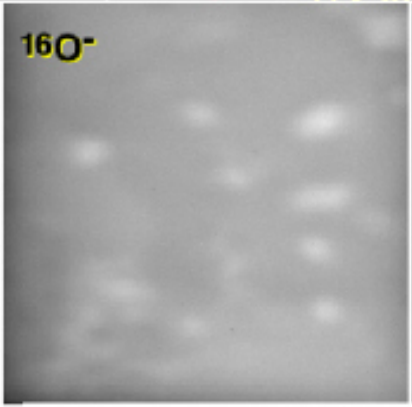


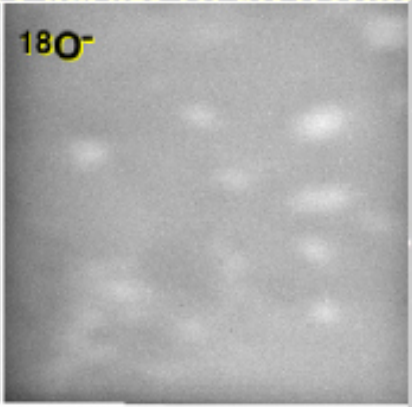


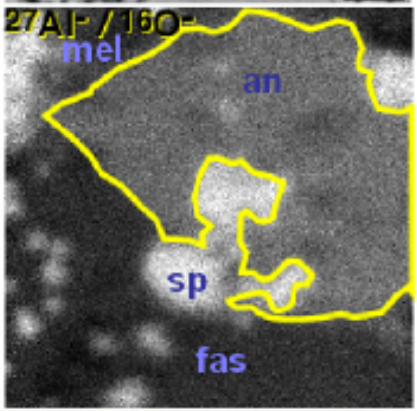
Application II: Oxygen isotopic variation in the early solar system

Nuclear Cosmochemistry
We are always on the frontier.



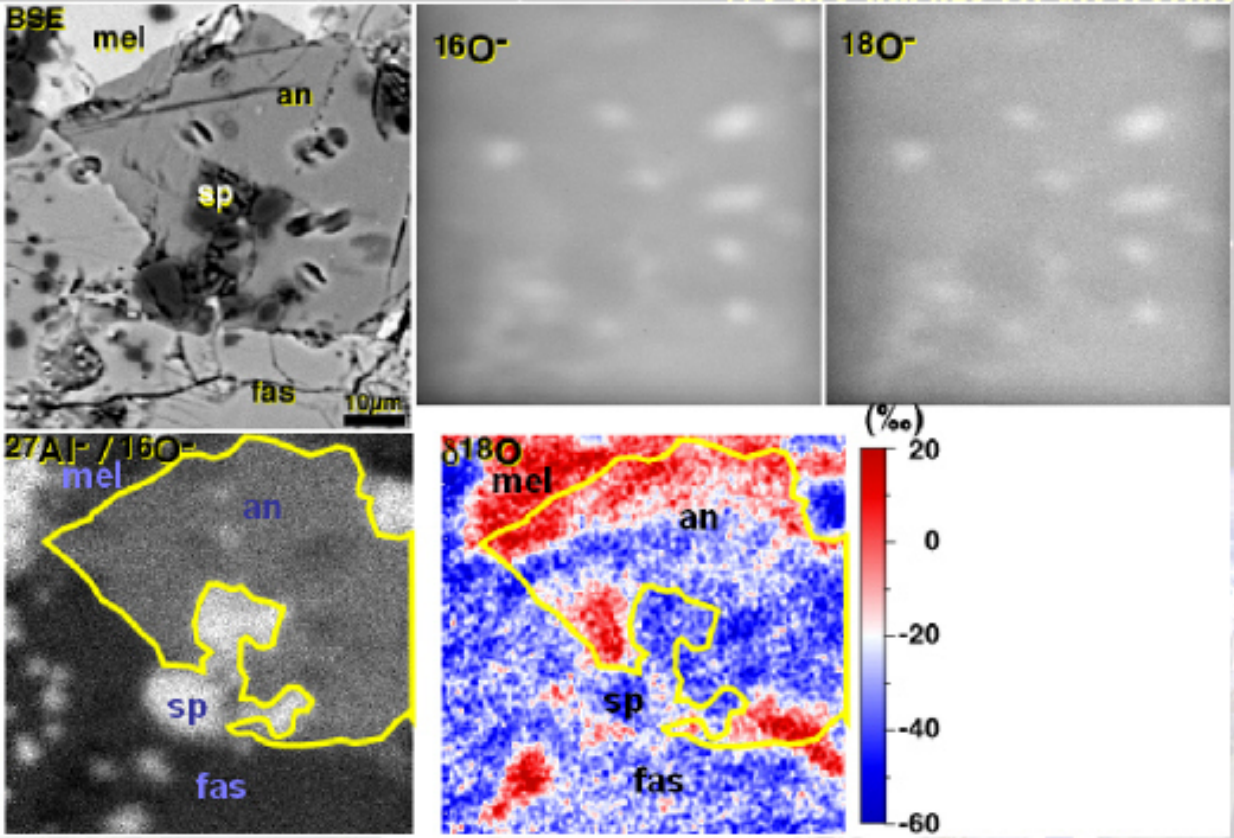






Application II: Oxygen isotopic variation in the early solar system

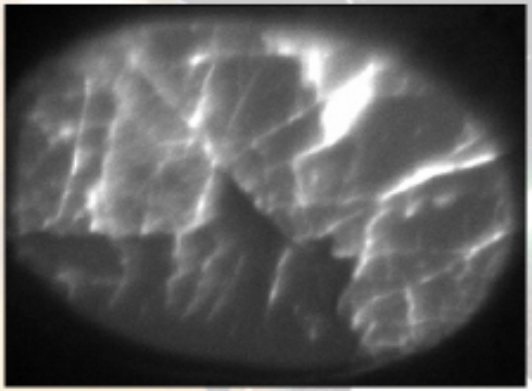
Nuclear Cosmochemistry
We are always on the frontier.



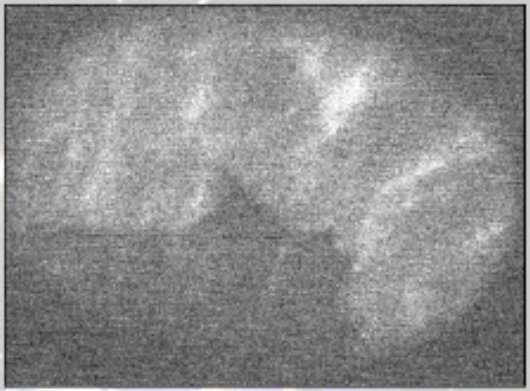
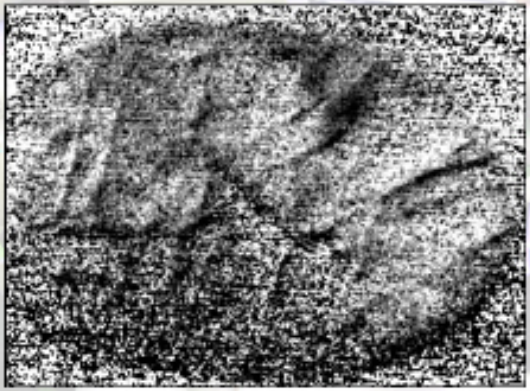
Application III: Hydrogen distribution and the isotopic variations of a Martian meteorite

Isotope Cosmochemistry
We are always on the frontier.

H: 99.9885% ~0.1%



D: 0.0115% ~10ppm



D/H